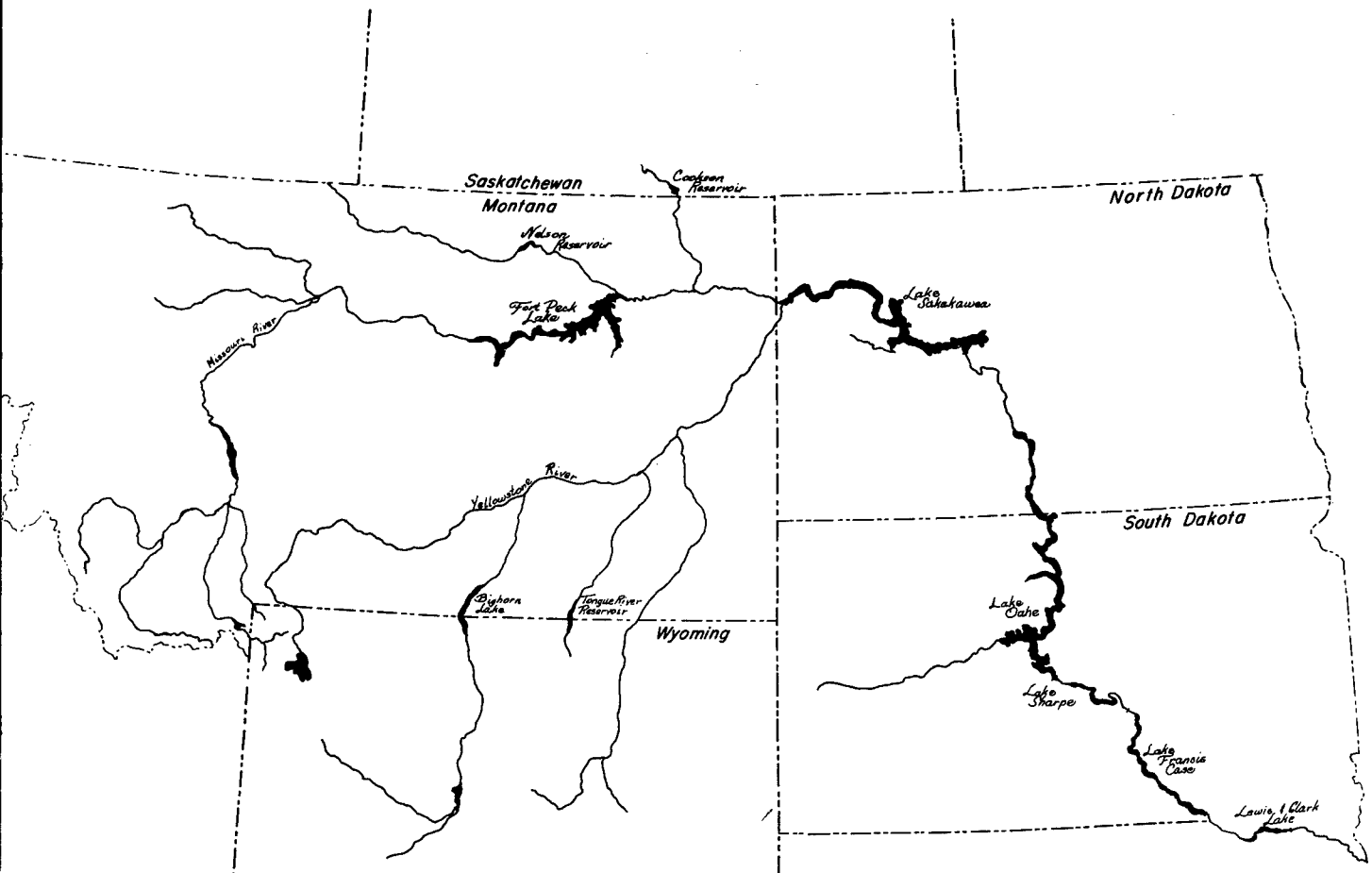
Factors Affecting the Mobilization, Transport, and Bioavailability of Mercury in Reservoirs of the Upper Missouri River Basin



UNITED STATES DEPARTMENT OF THE INTERIOR
FISH AND WILDLIFE SERVICE
Fish and Wildlife Technical Report 10

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By Glenn R. Phillips
Patricia A. Medvick
Donald R. Skaar
Denise E. Knight

Project Officers

Carl Armour
U.S. Fish and Wildlife Service

Denis Nelson
U.S. Environmental Protection Agency

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Factors Affecting the Mobilization, Transport, and Bioavailability of Mercury in Reservoirs of the Upper Missouri River Basin[1]

by

Glenn R. Phillips[2], Patricia A. Medvick,
Donald R. Skaar, and Denise E. Knight

Montana Cooperative Fishery Research Unit[3]
Montana State University, Bozeman, Montana 59717

Abstract

Factors controlling the mobilization, transport, and bioavailability of mercury in relation to coal mining and other mercury sources were studied in reservoirs of the Upper Missouri River Basin. We assessed mercury and selenium contamination of fishes and sediments in 10 reservoirs, estimated mercury fluxes in Tongue River Reservoir, determined dietary accumulation of methylmercury by fish, and related limnological conditions in three reservoirs to rates of mercury accumulation by fish. Detailed limnological studies were conducted in Nelson, Cookson, and Tongue River reservoirs. Mercury concentrations were higher in walleyes (*Stizostedion vitreum vitreum*) from headwater reservoirs with unregulated inflows than in fish of the same size from downstream reservoirs. Erosion and leaching during flooding apparently facilitated mercury accumulation by fish in reservoirs. Several observations led to this interpretation: (1)Northern pike (*Esox lucius*) in Tongue River Reservoir contained more mercury 1 year after a severe flood than in preceding or later years; (2) among fish of a given species and size, mercury concentrations were lower in fish from tailwaters than in those from the reservoir; and (3) turbidity, conductivity, total dissolved solids, nonfilterable solids, and pH were all strongly correlated with rate of mercury uptake by walleyes. About 93% of the mercury transported into Tongue River Reservoir was in river water. Point sources included 1% from mines and 9% from a sewage treatment plant; groundwater contributed only 0.02%, dry deposition 1%, and precipitation 4.5%. Nonpoint sources accounted for most of the mercury—emphasizing the importance of judicious land-management practices that help control erosion and leaching. Walleyes from Tongue River Reservoir fed chiefly on young-of-the-year white crappies (*Pomoxis annularis*), although young walleyes ate invertebrates in spring. White crappie diets varied diurnally: invertebrates were eaten primarily during daylight, and fish consumption increased at night. Total mercury in

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[2]Present address: Montana Department of Fish, Wildlife and Parks, Helena, Mont. 59620.
[3]The Cooperative Fishery Research Unit is jointly sponsored by the Montana Department of Fish, Wildlife and Parks, Montana State University, and the U.S. Fish and Wildlife Service.

forage organisms averaged 0.08 μg/g; calculated average concentrations of methylmercury in diets were 0.05 μg/g for walleyes and 0.04 μg/g for white crappies. Mercury concentrations in both species increased with increasing fish length and were higher in walleyes than in white crappies of the same age. The estimated percentages of methylmercury accumulated from food were 41–62 for walleyes and 51–73 for white crappies. Of the three reservoirs in which limnology was intensively studied, the rate of mercury uptake by fish was highest in Cookson Reservoir, followed by Nelson and Tongue River reservoirs. Characteristics of Cookson that facilitated mercury uptake included the relatively young age of the reservoir, high bacterial counts at the inflow, high water temperatures at the sediment–water interface, a low percentage of clay in the sediments, and the relatively high pH and conductivity of the water. Alternatively, factors contributing to the lower rate of mercury uptake by fish in Tongue River Reservoir included incomplete thermal mixing that resulted in cooler bottom waters, more reducing conditions near the bottom, more clay-like sediments, and higher concentrations of sulfide and oxides of iron and manganese in the sediments. Physicochemical conditions in some reservoirs seemingly enhance the bioavailability of mercury to fish, even in the absence of high mercury concentrations in sediments and water. Spillway design, basin shape, reservoir flow characteristics, watershed geochemistry, and other upstream conditions affect the vulnerability of a new reservoir to mercury problems. There is some opportunity to mitigate such problems because many of these factors can be controlled by judicious site selection, land management practices, and reservoir design and management.

Mercury concentrations exceeding former guidelines for human consumption established by the United States (1 μg Hg/g wet weight) or Canada (0.5 μg Hg/g wet weight) have been reported in edible flesh of fish from several western impoundments (the U.S. guideline was revised in 1978 to apply to methylmercury only). Examples include Lake Powell, Arizona (Potter et al. 1975); Antelope Reservoir, Oregon (Phillips and Buhler 1980); Tongue River Reservoir, Montana (Phillips and Gregory 1980); Southern Indian Lake Reservoir, Manitoba (Bodaly and Hecky 1979); Lahontan Reservoir, Nevada (Richins and Risser 1975); Cookson Reservoir, Saskatchewan (Waite et al. 1980); Lake Oahe, North Dakota and South Dakota (Walter et al. 1974); and Lake Fort Peck, Montana; and Lake Sakakawea, North Dakota (Nelson et al. 1977). Mercury lost during gold and silver milling operations is believed to have contributed to mercury in Lahontan and Antelope reservoirs and Lake Oahe; in the other impoundments, however, mercury probably originated from natural weathering.

Mercury is subject to interconversions in the environment between monomethylmercury (the predominant mercurial in fish tissue) and various less bioaccumulative inorganic chemical species (Wood et al. 1968). Although there is disagreement about whether these interconversions are biologically, chemically, or photochemically mediated (Jensen and Jernelöv 1969; Akagi et al. 1977; Rogers 1977), the upshot is that inorganic mercury can be converted in the environment to methylmercury. Fish, without themselves manifesting adverse effects, can accumulate methylmercury at concentrations that far exceed Federal guidelines for safe human consumption (McKim et al. 1976).

A considerable body of literature suggests that physical and chemical characteristics of water bodies largely determine rates of mercury methylation and subsequent bioavailability of mercury to fishes (D'Itri et al. 1971; Jackson and Woychuk 1980; Park et al. 1980). However, an understanding of the cycling of mercury in natural waters has been impeded by the lack of reliable techniques for measuring methylmercury concentrations as low as those present in all but a few of the most heavily contaminated waters (Miller 1977; National Academy of Sciences 1978; Park et al. 1980).

Laboratory studies have demonstrated that fish assimilate methylmercury from water across gill surfaces and from food by digestive absorption (Hannerz 1968; Lock 1975; Olson et al. 1975); mercury accumulated from the two sources is additive (Phillips and Buhler 1978). However, accounts in the literature about the relative importance of the two sources to fishes in natural waters conflict,

leaving unresolved the question of whether mercury is magnified through food chains. Methylmercury accumulation from water has not been directly quantified, because analytical techniques are not sensitive enough to detect the low concentrations occurring in most natural waters (Westöö 1975; National Academy of Sciences 1978). The alternative approach, direct quantification of methylmercury accumulated by fish from their food, has prove equally frustrating due to the difficulty of determining methylmercury consumption by fish (National Academy of Sciences 1978).

In 1978–1981, we studied factors controlling the mobilization, transport, and bioavailability of mercury in Upper Missouri River Basin reservoirs. Some of the mercury pathways examined are shown in Fig. 1. We were particularly concerned that land alterations and mining activities associated with energy development in the northern Great Plains might accelerate mercury dissolution, contributing additional mercury to reservoirs where some fishes already contained mercury concentrations exceeding governmental limits.

Accordingly, we (1) measured mercury and selenium concentrations in walleyes, surficial sediments, and sediment cores from 10 reservoirs in the Missouri River Basin and from portions of the Tongue River; (2) monitored mercury uptake trends in northern pike (*Esox lucius*) from the Tongue River Reservoir for 4 consecutive years; (3) compared rates of mercury uptake by river and reservoir fishes in the Poplar River–Cookson Reservoir and Tongue River Reservoir systems; (4) estimated influxes of mercury to the Tongue River Reservoir (surface coal mining, groundwater, domestic sewage, and atmospheric transport) and estimated downstream efflux and reservoir accumulation; (5) examined, for walleyes (*Stizostedion vitreum vitreum*) and white crappies (*Pomoxis annularis*), the food habits, food-consumption rates, mercury uptake from the diet, and total mercury and methylmercury concentrations in food organisms; and (6) related limnological variables in Nelson, Cookson, and Tongue River reservoirs to differences in rates of mercury uptake by walleyes in these waters.

These studies were conducted in portions of the Upper Missouri River Basin in Montana,

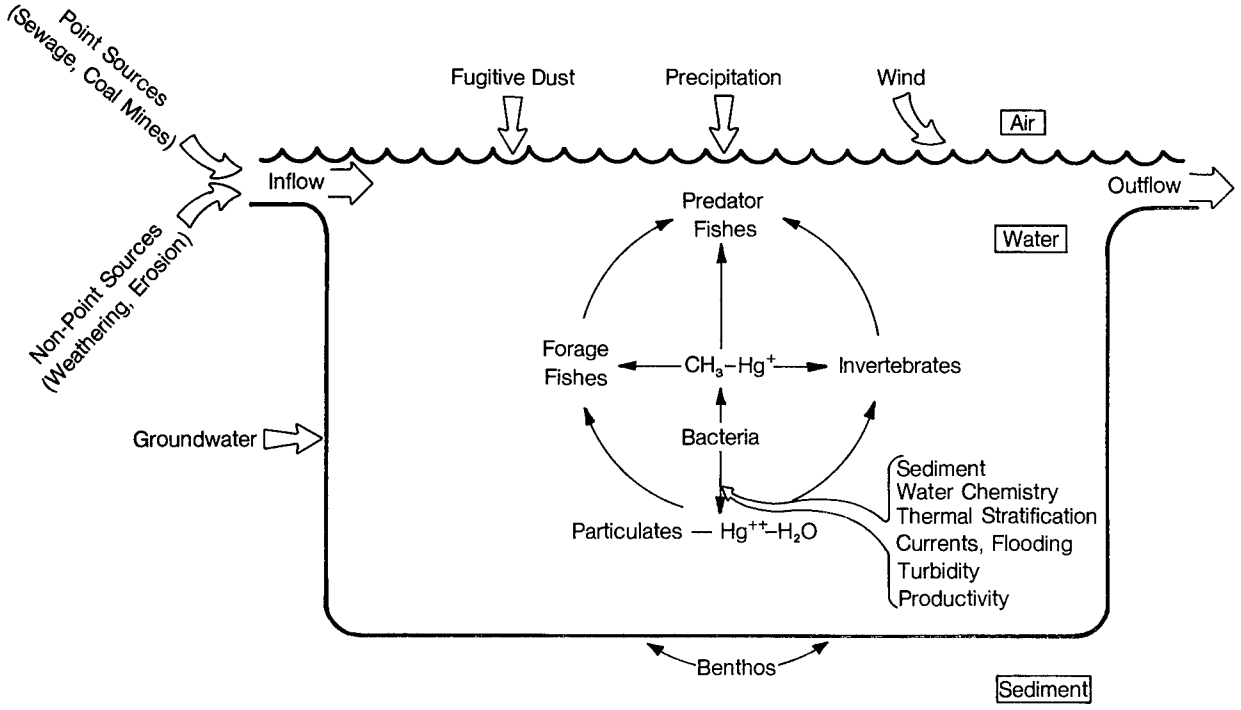

Fig. 1. Schematic showing sources of mercury to reservoirs and conditions influencing methylation of mercury.

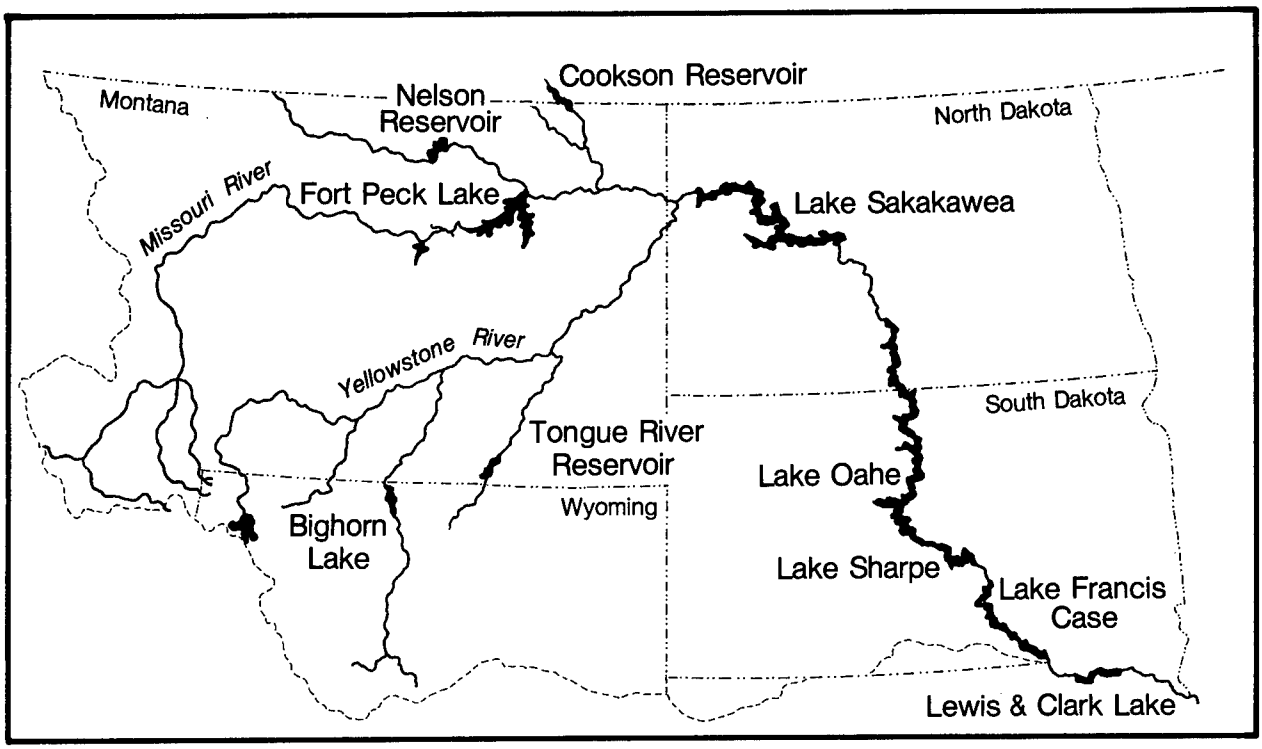

Fig. 2. Upper Missouri River Basin, showing the 10 reservoirs studied.

Wyoming, North Dakota, South Dakota, and Saskatchewan (Fig. 2). The walleye was the species of primary focus in most locations; however, a number of other species were collected: northern pike; common carp, *Cyprinus carpio*; golden shiner, *Notemigonus crysoleucas*; white sucker, *Catostomus commersoni*; white crappie; black crappie, *Pomoxis nigromaculatus*; yellow perch, *Perca flavescens*; and sauger, *Stizostedion canadense*. Other species mentioned in the text or tables are rainbow trout, *Salmo gairdneri*; Arctic char, *Salvelinus alpinus*; white bass, *Morone chrysops*; green sunfish, *Lepomis cyanellus*; largemouth bass, *Micropterus salmoides*; and threadfin shad, *Dorosoma petenense*.

Most of this work was done in Tongue River Reservoir, an irrigation and flood-control impoundment in southeastern Montana (Fig. 3). The reservoir is a mildly eutrophic and well-mixed hard-water impoundment (Whalen 1979). Important sport fishes include walleyes, white crappies, black crappies, saugers, and northern pike. Two other reservoirs that received considerable attention were Nelson Reservoir in north-central Montana (Fig. 4) and Cookson Reservoir in southeastern Saskatchewan (Fig. 5).

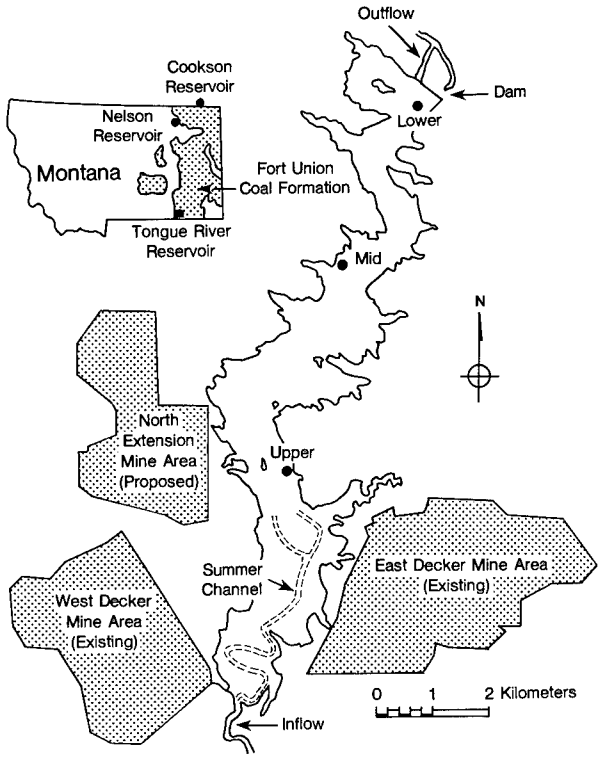

Fig. 3. Tongue River Reservoir, showing locations of the three sampling stations (upper, middle, and lower) for monitoring limnological characteristics and (*inset*) locations of Tongue River, Nelson, and Cookson reservoirs.

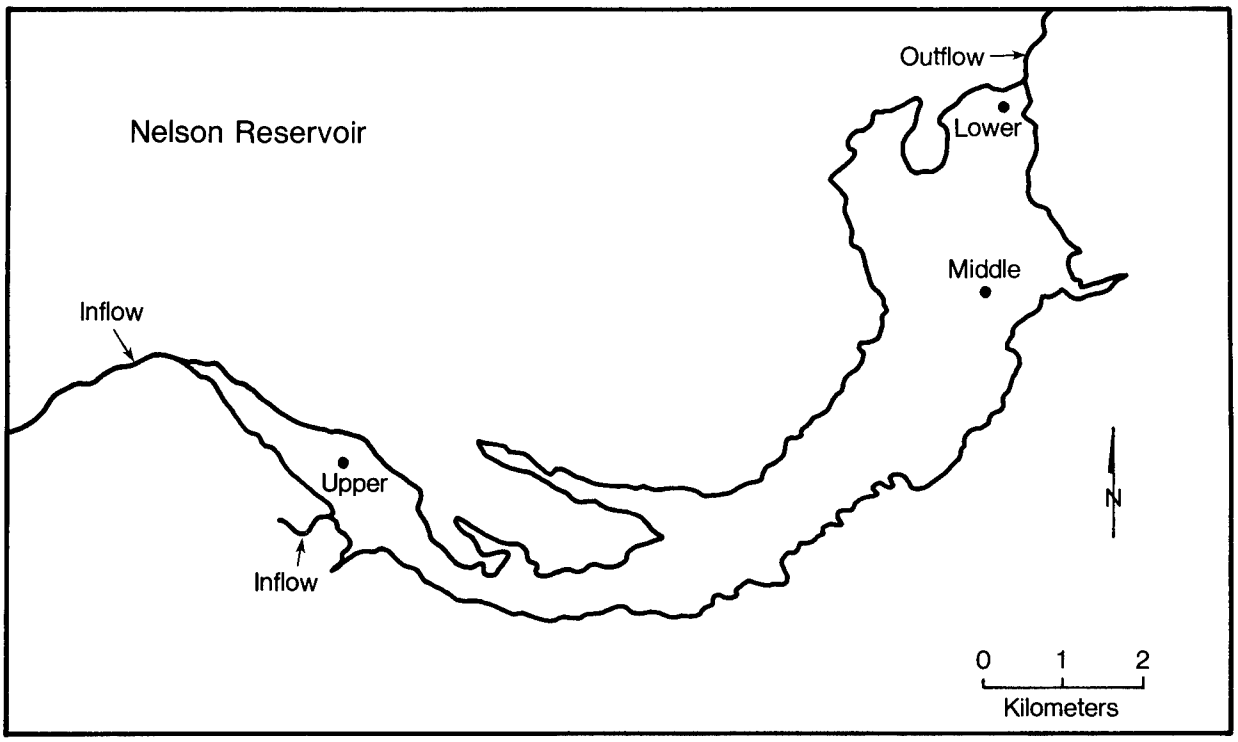

Fig. 4. Nelson Reservoir, showing locations of the three sampling stations (upper, middle, and lower) for monitoring limnological characteristics.

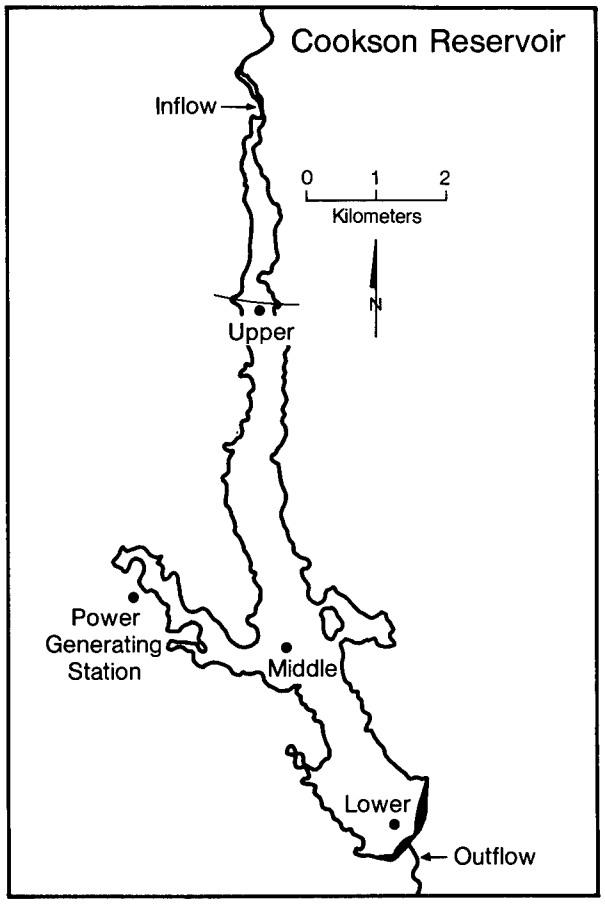

Fig. 5. Cookson Reservoir, showing locations of the three sampling stations (upper, middle, lower) for monitoring limnological characteristics.

Materials and Methods

Mercury and Selenium in Sediments and Fish from 10 Reservoirs

In our comparative assessment of mercury and selenium in sediments and fish from reservoirs of the Upper Missouri River Basin, we collected samples from 10 impoundments (Fig. 2): Tongue River Reservoir, Nelson Reservoir, Lake Fort Peck, Bighorn Lake, Cookson Reservoir, Lake Sakakawea, Lake Oahe, Lake Sharpe, Lake Francis Case, and Lewis and Clark Lake. Sediment samples and fish were also taken from the Tongue River and several branches of the Poplar River downstream from Cookson Reservoir. Earlier physical and chemical data pertaining to these reservoirs were summarized. In the absence of information on fish ages, we determined whether size and mercury were related in "standard" walleyes 500 mm long (as determined by regression of fish length against mercury content) from each location.

Mercury and Selenium in Sediments

Sediment samples were collected along two transects each in Lakes Sakakawea, Oahe, Sharpe, Francis Case, and Bighorn and along one transect in each of the other five reservoirs. Each transect extended from one shoreline across the reservoir to the opposite shoreline, perpendicular to the major direction of water flow. Exceptions were Lake Sakakawea and Bighorn Lake, where transects crossed large bays. Transects consisted of 10–30 evenly spaced surficial samples and one or two core samples taken near midchannel.

Surficial sediment samples were collected with an Ekman dredge, and sediment cores with a Phleger core sampler equipped with removable polycarbonate liners. Surface sediments were stored in sealed plastic bags, and core samples in the removable liners. All samples were placed on ice and later frozen. Preparation for metal analyses included thawing, homogenization, drying at 60 °C, and pulverization with a mortar and pestle. Core samples were divided into 5-cm sections for separate preparation and analysis. Concentrations of mercury and selenium in sediments are reported on a dry-weight basis.

Surficial sediments were taken from 16 locations in Goose Creek between Sheridan, Wyoming, and its confluence with Tongue River. We collected 42 samples in a section of Tongue River extending from about 1 km upstream from its confluence with Goose Creek to the upstream end of Tongue River Reservoir, and 17 samples downstream from the reservoir between Tongue River Dam and the town of Birney, Montana.

The walleye was the principal species for comparing rates of mercury uptake by fish in the various reservoirs because it was relatively abundant in all of the reservoirs except Lewis and Clark Lake. We sampled saugers instead of walleyes in Lewis and Clark Lake, at the request of the South Dakota Game, Fish and Parks Department. Fish were sampled from most of the reservoirs with gill nets fished overnight; graded mesh sizes were used in an attempt to obtain fish of a wide range of sizes. At Lake Fort Peck we collected walleyes with fyke nets, and at Nelson Reservoir we obtained tissue samples from fish caught by anglers. We sampled 47 to 100 walleyes from each impoundment except Nelson Reservoir (28) and Tongue River Reservoir (163). Common carp were collected from the Tongue River by electroshocking and from Tongue River Reservoir with gill nets. Walleyes were taken from the various branches of the Poplar River by electroshocking.

Northern pike were collected in Tongue River Reservoir, primarily with fyke nets and occasionally with gill nets, during spring for 4 consecutive years, 1978–1981. Most northern pike were marked with Floy anchor tags and returned to the reservoir after a muscle tissue sample had been surgically removed. The recovery of tagged fish that had been biopsied enabled us to monitor the uptake or elimination of mercury in individual fish. Over the 4 years of sampling, 17 northern pike were recaptured and biopsied during 2 or more years—including 1 in 3 of the 4 years, and 1 in all 4 years.

We took 20–50 g of axial muscle tissue in a dorsolateral area from all fish; tissue samples were placed in plastic bags, labeled, and frozen for later analyses of metal residues. Concentrations of mercury and selenium in tissue are reported on a wet-weight basis.

Analysis of Metals

Total mercury and selenium in sediments and fish tissues (discussed later) were determined with a Varian model AA-6 atomic absorption spec-

trophotometer equipped with a carbon rod atomizer, according to the method of Siemer and Woodriff (1974).

Analytical accuracy and precision were determined by analyses of known duplicates (2-5% of samples); blind duplicates (5-10% of samples); spiked samples (5-10% of samples); and U.S. National Bureau of Standards certified tuna (mercury in tissue), coal fly ash (mercury in sediment), and bovine liver (selenium in tissue). Quality control results for metal determinations are summarized in Table 1.

Statistics

The coefficients of determination (r^2) from regressions of fish length against mercury concentration in tissue (wet weight) were calculated after logarithmic transformation of mercury content. Regression slopes were compared by an F-test, and sample means were compared by using Scheffe's multiple comparison procedure (Neter and Wasserman 1974). Pearson correlation coefficients (r) were used in determining relations between mercury uptake by walleyes and the various physical,

Table 1. *Summary of quality control results for mercury and selenium analyses, including percent recovery of spikes and National Bureau of Standards reference samples and percent difference between duplicate samples.*

Sample[a]	n	Recovery (% variation from 100 %)[b]		Difference between duplicate samples (%)	
		Mean	SD	Mean	SD
Mercury					
Whole fish, wet weight					
Known duplicates	8			17.3	8.1
Spikes	21	13.7	9.3		
Tissue, wet weight					
Known duplicates	80			13.4	25.7
Blind duplicates	95			31.1	29.8
Spikes	268	11.4	8.5		
NBS tuna	40	11.0	8.8		
Sediment, dry weight					
Known duplicates	24			24.5	40.1
Blind duplicates	39			43.8	53.6
Spikes	76	12.8	9.5		
NBS coal fly ash	4	9.0	3.5		
Selenium					
Tissue, wet weight					
Known duplicates	19			21.6	32.5
Blind duplicates	54			25.4	21.3
Spikes	69	15.2	14.5		
NBS bovine liver	12	19.9	14.7		
Sediment, dry weight					
Known duplicates	19			23.1	29.2
Blind duplicates	25			140.4	163.6
Spikes	48	20.3	13.3		

[a]NBS = U.S. National Bureau of Standards.

[b]Difference between duplicate samples was calculated by using the formula $\left(\frac{\text{concentration of replicate } a}{\text{concentration of replicate } b} \times 100\right) - 100$, where the measured concentraion in sample a was greater than or equal to that in sample b.

chemical, biological, and temporal variables in the reservoirs. In comparing sample means we used Student's t-test (Steel and Torrie 1960). The accepted level of statistical significance is $P \leq 0.05$ unless otherwise specified.

Mass Balance Budget for Mercury in Tongue River Reservoir

To formulate a mass balance budget for mercury in Tongue River Reservoir, we measured mercury concentrations in water from Tongue River watershed twice monthly during April–September 1980 and monthly thereafter until February 1981. To determine point sources of mercury, we analyzed water discharged from several surface coal mines; and the sewage treatment plant in Sheridan, Wyoming. Most runoff from disturbed areas within the mine was collected and discharged from a point source. The Tongue River was sampled immediately upstream from the reservoir and in the reservoir discharge, as shown in Fig. 6 and described in Table 2.

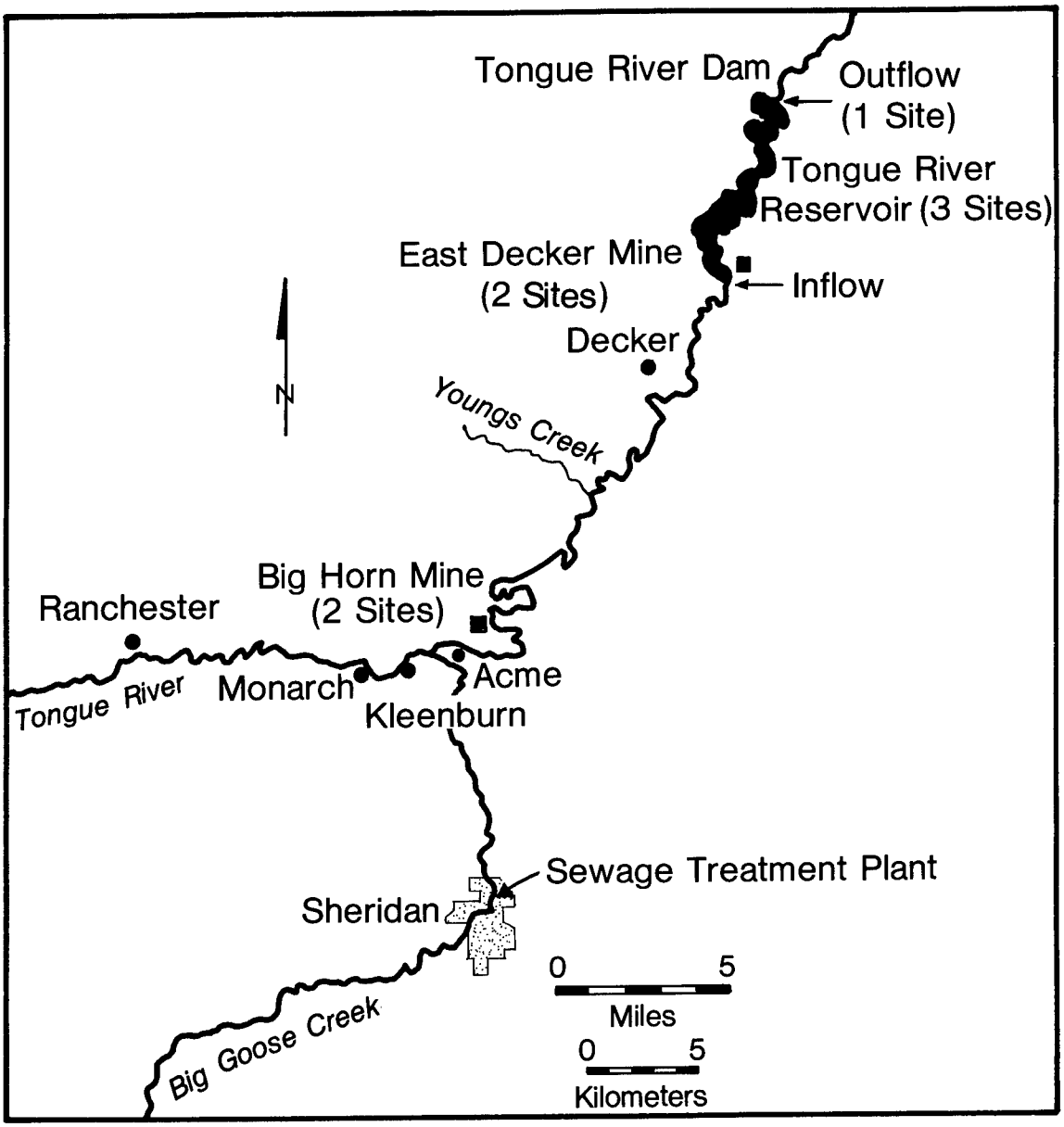

Fig. 6. Tongue River and Tongue River Reservoir showing locations of sampling stations for monitoring mercury content in water.

Table 2. *Descriptions of sampling sites, upstream to downstream, where mercury in water was monitored; see Fig. 6 for locations of sites.*

Sampling location	Description
Sheridan sewage outfall	Discharge channel from the Sheridan sewage treatment plant (empties into Goose Creek).
Bighorn Mine	
Upper	Discharge at plume in channel from upper settling pond.
Lower	Discharge at plume in channel from lower settling pond.
East Decker Mine	
South	Discharge at plume in channel from south settling pond.
North	Discharge at plume in channel from north settling pond.
Tongue River Reservoir[a]	
Inflow	Tongue River below bridge to East Decker Mine
Station 1	Upstream end of Tongue River Reservoir
Station 2	Midlocation in Tongue River Reservoir
Station 3	Downstream end of Tongue River Reservoir
Outflow	Tongue River about 100 m downstream from Tongue River Dam

[a]At stations 1, 2, and 3 samples were taken at surface, middepth, and bottom. See Fig. 3 for locations.

Water collected for total mercury analyses was placed in 250-mL glass bottles sealed with Teflon-lined caps, and preserved in potassium dichromate (0.05%) and concentrated nitric acid (1.5 mL/L). Samples were iced immediately after collection and analyzed for mercury within 21 days by atomic absorption spectrophotometry (Siemer and Hageman 1980). Quality-control activities included analyses of blind duplicate samples (monthly) and blind spiked samples (quarterly). Reagent blanks were also analyzed with each batch of samples.

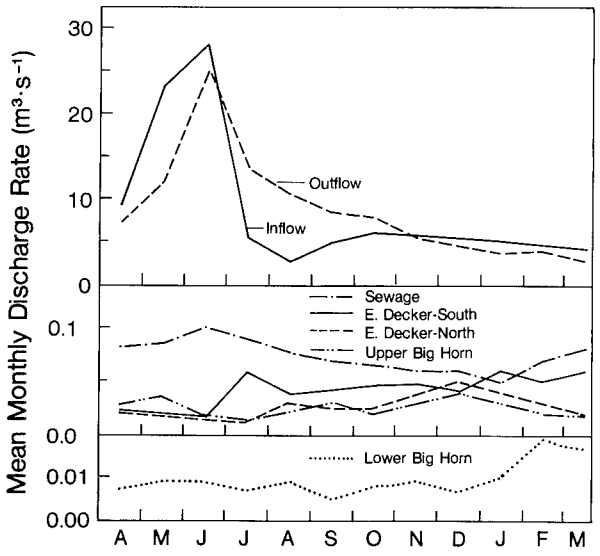

Fig. 7. Average monthly flow rates at water sampling sites in inflow and outflow of Tongue River Reservoir and at other sites (see Fig. 6 for locations).

Tongue River flows (Fig. 7) were obtained from the U.S. Geological Survey (USGS); effluent flow rates were provided by the appropriate agencies. Records of precipitation, intermittent streamflow, and seepage rate of groundwater were obtained from the literature (Table 3). Information on mercury concentrations in groundwater near the reservoir in 1980 and 1981 was provided by Hittman Associates, Inc. (1981*a,b,c*).

Annual mercury discharges or loadings for Tongue River and effluent from Sheridan sewage treatment plant and coal-mine ponds were estimated by multiplying average mercury concentrations for each site and month (for February and March we used overall average mercury concentration for the location) by the average monthly flow rates (Table 4). The monthly flux of mercury at each site was obtained simply by multiplying by the appropriate constant. Sums of the values for each site yielded total grams of mercury moving past each sampling location per year.

For intermittent streams, groundwater, and precipitation, average mercury concentrations were multiplied by the annual water input rates from each source to obtain annual mercury load into the Tongue River or Tongue River Reservoir.

Table 3. *Data and literature sources for the mercury budget for Tongue River Reservoir.*

Component	Mercury		Flow	
	Mean concentration (μg/L)	Source	Rate (L/year)	Source
Intermittent streams	0.01	Present study	5.60×10^9	Rykiel and Hall 1978
Groundwater	0.057	Hittman Associates, Inc. 1981*a*	1.23×10^7	Rykiel and Hall 1978
Precipitation	0.05	Skogerboe et al. 1980	3.80×10^9[a]	USGS; Hittman Associates, Inc. 1981 *a*,*b*, and *c*.
Dry deposition	0.034[b]	Present study	34.6[c]	Decker Coal Co.
Inflow[d]	0.015	Present study	2.75×10^{11}	Present study, 1980–81
Outflow[d]	0.018	Present study	2.69×10^{11}	Present study, 1980–81
Reservoir	0.017	Present study	8.55×10^{10}	Rykiel and Hall 1978

[a]Reservoir surface area multiplied by 299.5 mm/year.
[b]Dust, μg Hg/g.
[c]Particulates in air, $\mu g/m^3$.
[d]Calculations were based on monthly averages.

Table 4. *Average and (in parentheses) range of mercury concentrations and water flow rates for monitored sources and sinks of mercury in Tongue River Reservoir, and resultant mercury fluxes during April 1980–March 1981.*

Station	Mercury concentration (μg/L)	Waterflow rate (m^3/s)	Mercury flux (grams)	
			Daily	Monthly
Sheridan sewage effluent	0.16 (0.10–0.30)	0.07 (0.05–1.00)	1.05 (0.48–2.31)	32.1 (14.7–40.2)
Bighorn Mine				
Upper pond	0.01 (0.01–0.03)	0.03 (0.01–0.04)	0.03 (0.01–0.10)	1.0 (0.3–3.2)
Lower pond	0.02 (0.01–0.06)	0.01 (0.01–0.02)	0.02 (0.01–0.04)	0.5 (0.2–1.1)
East Decker Mine				
South pond	0.01 (0.01–0.02)	0.04 (0.02–0.06)	0.05 (0.02–0.05)	1.5 (0.6–2.5)
North pond	0.01 (0.01–0.03)	0.02 (0.02–0.05)	0.03 (0.02–0.09)	0.9 (0.7–2.7)
Tongue River Reservoir				
Inflow	0.01 (0.01–0.03)	8.8 (2.9–28.2)	11.1 (2.4–40.7)	335 (75–1,220)
Outflow	0.02 (0.01–0.04)	8.9 (2.8–24.4)	14.9 (3.2–51.3)	453 (134–1,538)

The quantity of mercury present in the reservoir water was calculated by multiplying the average mercury content of the water (0.014 μg/L) by the volume of the reservoir at full pool (supplied by USGS).

We collected 28 surface dust samples in April 1981 from several locations near the reservoir (Fig. 8) and analyzed them for mercury by atomic absorption spectrophotometry (Siemer and Woodriff 1974). Average annual concentration of total solid particulates in air was obtained from several high-volume air-sampling devices at East Decker and West Decker mines. The average mercury concentration in dust (0.034 μg/g) and the average concentration of particulates in air were used to estimate dry deposition of atmospheric mercury.

Dry deposition was estimated by the following formula, modified from Kramer (1978) and Brzezinska and Garbalewski (1980).

$$D_{TRR} = C_A V_d S_{TRR}$$

where

D_{TRR} = dry deposition rate to Tongue River Reservoir,

C_A = aerosol concentration (μg Hg/m^3),

V_d = deposition velocity (1 cm/s), and

S_{TRR} = surface area of Tongue River Reservoir (12.8 km^2).

Deposition velocity (V_d) varies with particle size, wind velocity, and depositional interface. A value of 1 cm/s is often used to calculate particulate deposition on surfaces (Skogerboe et al. 1980). A constant (3.1536 m/g per year) was used to transform the value to grams of Hg per year.

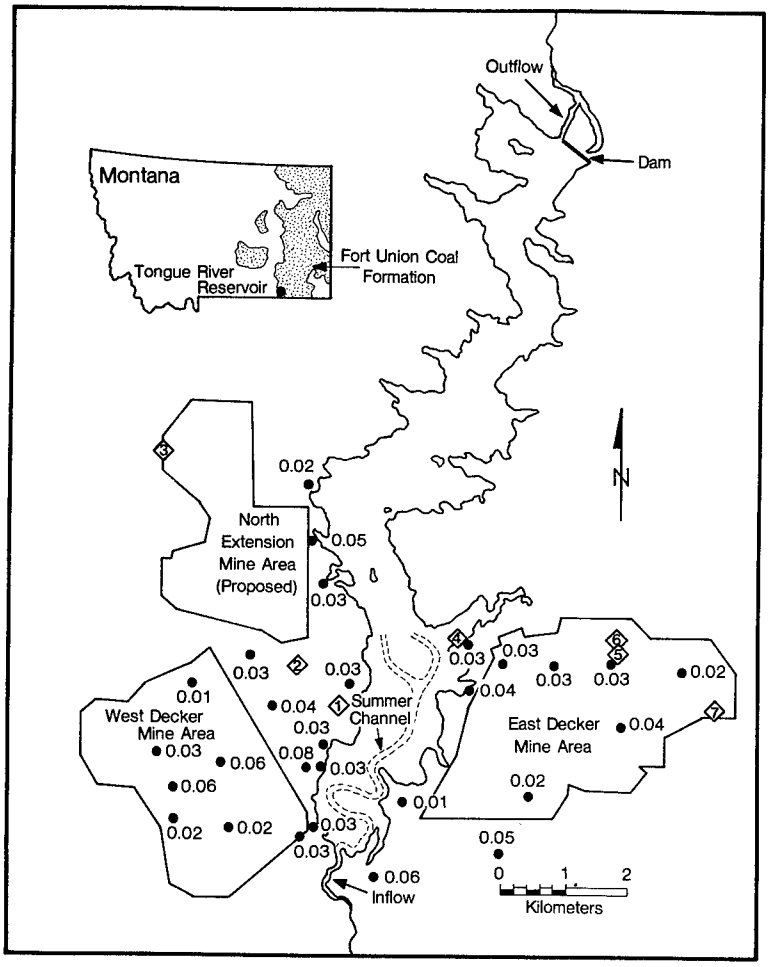

Fig. 8. Collection locations for surface dust samples (*dots*) and mercury concentrations (μg/g) in dust. Diamonds enclosing numbers 1 to 7 show locations of high-volume air-sampling devices monitored by Decker Coal Company.

Accumulation of Dietary Methylmercury by Fish

Field Collections

In studies of dietary accumulation of methylmercury by fish, we collected walleyes and white crappies and their food organisms at 2-month intervals from the time of ice breakup in April through October 1980. Sampling was confined primarily to the middle third of the reservoir, since Riggs (1978) showed on the basis of mark–recapture data that both species move freely throughout Tongue River Reservoir.

White crappies were collected at 3-h intervals in single-lead trap nets. Samples were taken at all times of day, and sets were repeated if the sample size for a given time interval was small. The short sampling intervals enabled us to observe daily feeding peaks and provided almost completely undigested stomach contents. Walleyes were collected with single- and graded-mesh gill nets fished overnight. Fish from these nets yielded identifiable stomach contents, thus eliminating the need for more frequent sampling. Forage fish were sampled with both gill nets and trap nets, and invertebrates with sweep nets and an Ekman dredge.

We tried to collect 30 to 50 walleyes and 60 to 80 white crappies (6 to 10 crappies for each 3-h sampling interval) during each of the four sampling periods (April, June, August, and October). The catch was subsampled to include a wide size range. All fish were sacrificed, weighed to the nearest 10 g, and measured (total length) to the nearest 1 mm. A muscle sample was taken from the anterior dorsal section of a fillet of each fish and frozen for later mercury analysis. The stomachs of walleyes and the entire gastrointestinal tracts of crappies were removed, preserved in 70% ethanol, and stored for later identification of the contents.

Stomach contents were periodically inspected in the field, and predominant food organisms were collected and frozen whole for mercury analysis and estimation of mercury in the fish diet. For comparison the frozen stomach and intestinal contents of 80 white crappies (representing all sampling periods except June) were also analyzed for mercury. A few (5 to 10) walleyes and white crappies were homogenized entirely, allowing us to compare mercury concentrations in whole fish with concentrations in muscle.

Mercury Analysis

Total mercury concentrations (μg/g) in fish and invertebrates were measured with a Varian model AA-6 atomic absorption spectrophotometer by the method of Siemer and Woodriff (1974). Whole fish were homogenized in a blender with dry ice and a subsample of the resulting frozen powder was analyzed for total mercury. The system was calibrated with freshly mixed standard solutions and tissue samples of known mercury concentration. Blind and known duplicate analyses of samples were also performed. Mean percent difference was 13.4 (0.03 μg Hg/g) for known tissue duplicates, 17.3 (0.01 μg Hg/g) for known whole-fish duplicates, and 31.1 (0.08 μg Hg/g) for blind tissue duplicates. All mercury concentrations are reported on a wet-weight basis. Concentration of methylmercury (MeHg) in whole fish was determined by the method of Watts et al. (1976).

Food of Walleyes and White Crappies

Volumes of stomach contents and (for crappies) intestinal contents were measured to the nearest 0.05 cm^3 by displacement in a calibrated centrifuge tube; stomach contents of white crappies were weighed to the nearest 0.01 g. Fish found in stomachs were identified to species, and their individual lengths and volumes measured when possible. Invertebrates were identified to order or family, depending on the state of digestion. Total volumes of invertebrates were measured directly for each stomach; percent volumes were estimated individually for orders and families with volumes too small for direct measurement. Percent volume and percent frequency of occurrence were calculated for major dietary components by season (sampling period), length, and time of day (crappies only). Fish were divided into length categories according to growth rates and length- and age-group estimates for walleyes (Riggs 1978) and white crappies (Elser et al. 1977) from Tongue River Reservoir. Live total lengths of prey fish eaten were estimated from their partly digested remains; linear regression equations (Snedecor and Cochran 1967) were used to estimate the total lengths of partly digested fish based on body proportions of undigested fish (Table 5).

Table 5. *Linear regression equations used to estimate total length (mm) of ingested prey fish from portions of their digested remains. Equations were derived from a series of length measurements made on whole fish (P < 0.001 for all equations).*

Forage species and equation for estimating total length in millimeters	r^2
Crappies	
1.30 (standard length) + 1.65	0.99
1.67 (trunk and operculum) + 1.39	0.99
1.93 (trunk length) +2.72	0.99
Golden shiner	
1.33 (standard length) − 3.71	0.99
1.53 (trunk and operculum) − 2.77	0.99
1.69 (trunk length) − 3.21	0.99
Yellow perch	
1.24 (standard length) − 4.68	0.90
1.47 (trunk and operculum) + 1.64	0.88
1.73 (trunk length) − 3.02	0.89
All Species combined	
1.19 (standard length) + 6.27	0.94
1.41 (trunk and operculum) + 12.83	0.92
1.60 (trunk length) + 13.62	0.92

Food-consumption Rates

Walleyes. We estimated annual food-consumption rates of walleyes in Tongue River Reservoir for each size-class from specific growth rates (g/g fish per day) and metabolic requirements, using the bioenergetics model of Kitchell et al. (1977), as modified by Breck and Kitchell (1978). Metabolic requirements were predicted from average body weights and average reservoir temperatures. Age and growth data for walleyes in Tongue River Reservoir (Riggs 1978) were used to estimate specific growth rates and average body weights. The year was divided into a growing period (May–September) when average monthly reservoir temperatures exceeded 12 °C (the physiological threshold for growth of walleyes as reported by Kelso 1972), and a nongrowing period (October–April) when monthly temperatures were below 12 °C. Over several years the average temperature was 18.2 °C for the growing period and 4.7 °C for the nongrowing period (Whalen 1979; Leathe 1980). Consumption estimates for the two periods were averaged to obtain an annual ration (R). Multiples of the standard metabolic rate of a species—commonly referred to as Winberg I, II, and III—were used to estimate resting, average, and maximum metabolic rates (activity levels) of walleyes (Winberg 1956; Ware 1975). Possible combinations of these activity levels for growing and nongrowing periods gave a range of annual consumption values for use in estimating the uptake of methylmercury (MeHg).

White crappies. We estimated food-consumption rates of white crappies for each sampling period from daily feeding peaks, using the field method described by Nakashima and Leggett (1978). Size-classes were pooled to increase sample size. For each 3-h sampling interval, the total wet weights of the digestive tract contents (stomach plus intestine) were corrected for the effects of preservation, pooled by sampling period, and expressed as a percentage of the total weight of the fish. Graphs of these values plotted against time showed feeding peaks, which were summed to provide an estimate of 24-h food consumption for that month. Consumption rates for May, July, and September were estimated by extrapolating between calculated values. Maintenance rations based on the estimate of Kitchell et al. (1977) for 100-g yellow perch, at mean monthly reservoir temperatures, were assumed for November–March.

Monthly estimates were summed to obtain an annual estimate, and standard errors were calculated (Snedecor and Cochran 1967), providing a range of consumption values for use in calculating MeHg uptake from food.

Methylmercury in the Diet

Methylmercury concentrations in the diet were calculated for each size-group of walleyes and white crappies. Total mercury concentrations in food items were measured directly, whenever possible, and converted to MeHg concentrations by multiplying by the percentage of total mercury present as MeHg, as reported in the literature (Knight 1982); literature values for MeHg were 6–76% (average 33%) for invertebrates and 6–100% (average 85%) for fish (usually muscle). Because of the wide range of values reported, we used the low, mean, and high MeHg percentages in calculations.

Diets were divided into invertebrate and fish components to estimate MeHg concentrations. Invertebrates were subdivided by order; orders composing 3% or less of the total stomach volume of fish of a given size-group were combined. Forage fish were subdivided by species into 10-mm length intervals, and the mercury concentration in fish from each interval was estimated from species regression equations (Snedecor and Cochran 1967) for concentration against total fish length (Table 5). The fraction of the diet represented by each component was then multiplied by the appropriate MeHg concentration and summed, giving the overall concentration in the diet.

Observed Accumulations

Methylmercury accumulation rates were determined by comparing regression equations for total mercury concentration against fish length for different years. Average mercury concentrations of fish in each size-group were estimated for 1978 and 1980 from estimated lengths of these fish in each year; multiplication by the average weight then gave the average amount of total mercury present in the fish in that year. The difference in these amounts, divided by 2, was the annual total mercury accumulation rate. This value was multiplied by the percentage of total mercury present as MeHg in fish, giving the anuual rate of MeHg accumulation (*dM/dt*). Low, mean, and high percentages of MeHg were used to calculate *dM/dt*, giving values designated as *dM/dt*—low, *dM/dt*—mean, and *dM/dt*—high.

Uptake from food. The accumulation of MeHg from food was calculated from the equation

$$\frac{dF}{dt} = a\,R\,C\,W, \qquad (1)$$

where *dF/dt* is the rate of MeHg uptake from food in μg/year, *a* is the assimilation efficiency of MeHg from the diet (percent × 0.01), *R* is the yearly ingested ration (g/g), *C* is the concentration of MeHg in diet (μg/g), and *W* is the average weight of fish in that size-class (g). This equation is similar to that used by Norstrom et al. (1976) to model the food-uptake component of MeHg accumulation by fishes.

Calculations for mean, high, and low values of *R* and *C* have already been described. For *a*, a wide range of values (15–88%) was found in the literature. Both extremes have been used in previous models (Fagerström and Åsell 1973; Norstrom et al. 1976); consequently, the two extremes and a mean value (33%) were used in our calculations. Of the 27 possible values for *dF/dt*, 5 were calculated for each size-class of walleyes and white crappies. Maximum and minimum values provide a possible range for *dF/dt*, and the mean values, from the means of *a*, *R*, and *C*, represent the middle ground. The lowest assimilation efficiency reported is apparently the most accurate under natural conditions (Phillips and Gregory 1979); consequently, we also calculated mean and maximum values of *dF/dt*, assuming the minimum value for *a*, and designated them as the low-mean and low-high values for *dF/dt*.

Fraction attributable to food. Methylmercury uptake from food (*dF/dt*) was compared with observed MeHg accumulation (*dM/dt*) in two ways. A crude estimate of the fraction derived from food (*FF*) was obtained by summing *dF/dt* over age and dividing by predicted 1980 mercury levels for each size-group of fish. With this method it is assumed that all accumulated mercury is MeHg, and no correction is made for elimination.

A more rigorous procedure for estimating the fraction derived from food involves correction of *dM/dt* for MeHg eliminated over the course of the year. Most investigators (Järvenpää et al. 1970; Miettinen 1975; Huckabee et al. 1979) have found MeHg elimination to be an exponential decay or half-life function. Half-life values in the literature range from 0.3 to 7.0 years and average 2.25 years (Knight 1982). Roughly equivalent annual elimination rates (*dE/dt*) are 10–90% (range) and 30% (average) of the body burden. Although most reported half-lives are near the mean, many are probably underestimates, because they were derived from very short tests during which little or no elimination occurred. The *FF* values were therefore calculated from both mean and low elimination rates (*dE/dt*—mean and *dE/dt*—low); high elimination rates, which were generally reported for trout or small fishes, were not used.

The fraction of the total MeHg accumulation derived from food (*FF*) was then calculated from the equation

$$FF = \frac{dF/dt}{dM/dt} = dE/dt\ (0.5\ dM/dt). \qquad (2)$$

All symbols are as previously defined. The fraction of total accumulated MeHg derived from

water was assumed to be (1 minus *FF*). In these calculations we used absolute amounts of MeHg in fish, rather than concentrations, to compensate for the effects of growth.

Relation of Reservoir Limnology to Mercury Accumulation by Fish

We studied the limnological characteristics of three Missouri River reservoirs—Tongue, Cookson, and Nelson—in relation to rates of mercury accumulation in fish. Three stations (upper, middle, and lower) in each reservoir (Figs. 3-5), and the inflows and outflows, were sampled monthly from April through October 1981. Dissolved oxygen, temperature, pH, specific conductance, and redox potential (E_h) were measured at 2-m depth intervals at each reservoir station with a Hydrolab Model 8000 water quality analyzer (Hydrolab Corporation, Austin, Texas), calibrated before and after each day's use according to the manufacturer's suggested procedures. Results for temperature and pH were compared periodically with those from a calibrated mercury thermometer and pH meter.

Water samples for determining density of bacteria were obtained with a Kemmerer sampler at mid-depth and bottom at each reservoir station and with glass bottles at the surface and at the inflows and outflows of each reservoir. At the time of collection, samples were preserved in 2 % formalin and iced. In the laboratory, samples were diluted, stained with acridine orange, and filtered (Hobbie et al. 1977); bacteria were counted with a Leitz epifluorescent microscope.

Initially, water for total mercury analysis was obtained at the intakes and outflows and at the surface, mid-depth, and bottom at each station; however, because mercury concentrations in this and previous studies (Phillips 1979) were consistently near detection limits (0.01-0.03 μg/L), sampling was limited to the inflow and outflow, and mid-depth at the middle station of each reservoir. We used a Van Dorn sampler to collect water at depth, and samples were processed and analyzed as described earlier. Accuracy of analyses was verified by known duplicates (5-10% of samples), blind duplicates (5-10%of samples), and spike and recovery with water samples certified by the U.S. Environmental Protection Agency (EPA). In the range of concentrations encountered, we estimated precision to be ±0.01 μg Hg/L.

In April and May 1981, one sample of benthic invertebrates was taken at each station with an Ekman dredge (0.0232 m^2). Invertebrates were removed from the sediments by sieving through a 0.6-mm mesh screen and were preserved in 10% formalin.

Sediment samples for chemical and physical analyses were obtained with an Ekman dredge in May 1981 and immediately frozen on dry ice. Before analysis the sediments were thawed, dried at 35-50 °C, and pulverized with a mortar and pestle. Samples for total iron and manganese were digested in a perchloric, nitric, and hydrofluoric acid solution. Extractable iron and manganese were removed by shaking for 2 h in 0.1 *M* NH_2OH:HCl/0.01 *M* HNO_3 (pH = 2) and filtering (Jackson and Woychuk 1980). Concentrations of Fe and Mn were determined by flame atomic absorption spectrophotometry (Crock and Severson 1980). Samples for total sulphur were digested with nitric, perchloric, and phosphoric acids, precipitated with barium to form barium sulphate, and turbidimetrically determined for concentration (Sulphur Institute 1968). Samples for total phosphorus were dry-ashed at 600 °C for 4 h and digested in 1:3 hydrochloric acid. Concentrations were determined with a colorimetric autoanalyzer (Black et al. 1965*b*). Total nitrogen samples were digested in sulfuric acid and determined by indicator titration (Black et al. 1965*c*). Percent ash weight was established by drying the sample at 60 °C for 24 h and ashing it at 600 °C for 4 h in a muffle furnace. Particle size was determined by the hydrometer method for mechanical analysis (Black et al 1965*a*).

Results and Discussion

Mercury and Selenium in Sediments and Fish from 10 Reservoirs

Reservoir Characteristics

The physical characteristics and age of the reservoirs of the Upper Missouri River Basin differed considerably (Table 6). Volumes ranged from 30.2 × 10^9 m^3 for Lake Sakakawea to only 0.04 × 10^9 m^3 for Cookson Reservoir, and maximum

Table 6. *Physical characteristics of Missouri River Basin Reservoirs.*[a]

Reservoir	Volumn[b] ($\times 10^9$ m^3)	Surface[b] area (km^2)	Depth (m)[b] Mean	Depth (m)[b] Maximum	Outflow height (m from bottom)	Age[c]	Theoretical average water retention time (years)
Sakakawea	27.7	1489	14.7	58.2	31	28	1.3
Oahe	27.4	1441	11.9	62.5	38	23	0.9
Fort Peck	22.1	971	16.3	67.1	40	44	1.7
Francis Case	5.7	384	11.2	44.2	27	29	0.2
Sharpe	2.2	227	1.3	23.8	23	18	0.1
Bighorn	1.8	73	24.0	140.0	76	16	0.3
Lewis and Clark	0.5	113	1.7	17.7	11	26	0.02
Tongue River	0.08	13	5.9	18.0	3	42	0.10
Nelson	0.07	19	4.0	14.2	0	65	0.19
Cookson	0.04	7	5.6	14.0	3	6	2.9

[a]Unpublished data of U.S. Army Corps of Engineers.
[b]Maximum normal operating pool elevation.
[c]Number of years before 1981 when dam was closed.

depths from 140 m in Bighorn Lake to 14 m in Cookson. Water was withdrawn from the bottom of most of the impoundments, exceptions being Bighorn, Sakakawea, and Fort Peck lakes (Table 6). Reservoir ages (in 1981) ranged from only 6 years for Cookson to 65 years for Nelson; average water retention times varied from 0.02 year for Lewis and Clark to almost 3 years for Cookson.

Turnover rates and flow regimes also varied among reservoirs. In Cookson, Nelson, Bighorn, and Tongue River reservoirs and the Big Dry Arm of Lake Fort Peck, the fluctuations in turnover frequency were extreme, the peak turnover rate occurring in spring or early summer; in the other reservoirs the annual flow regimes were less variable (Fig. 9). Inasmuch as flow regimes are a function of the amount of flow regulation upstream from each reservoir, headwater reservoirs are subject to the most extreme fluctuations in flow.

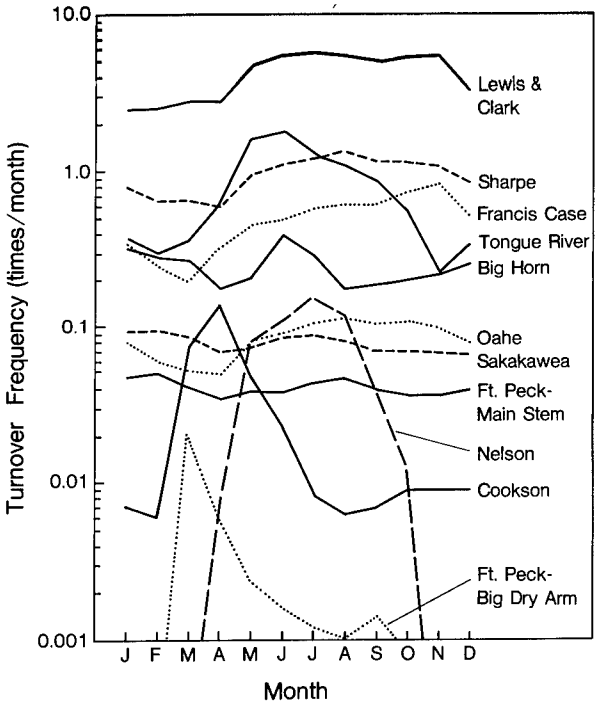

Fig. 9. Turnover frequencies of Missouri River Basin reservoirs. Data for the Big Dry Arm of Lake Fort Peck are based on the assumption that this arm acts as an independent water body. Monthly data represent averages over the periods for which records have been kept.

Sediments

Mercury concentrations in reservoir sediments were low (Table 7) relative to those at other locations (Table 8). Mean concentrations ranged from 0.02 μg/g at the upstream location in Lake Francis Case to 0.07 μg/g in both the Big Dry Arm of Lake Fort Peck and the downstream location in Lake Sharpe. Mercury in sediments was significantly and positively correlated with depth at 10 of the 14 sampling locations (Table 9); correlations were

Table 7. *Mercury and selenium concentrations in surficial sediments (top 5 cm) from 10 reservoirs in the Missouri River Basin.*

Reservoir and sampling location	Distance of site from inflow (km)	No. of samples	Water depth (m)			Mercury concentration (μg/g dry weight)			Selenium concentration (μg/g dry weight)		
			Mean	Range	SD	Mean	Range	SD	Mean	Range	SD
Tongue River											
Upper[a]	3.6	20	-	-	-	0.04	0.01-0.06	0.01	-	-	-
Lower[a]	11	23	-	-	-	0.04	0.01-0.07	0.02	-	-	-
Bighorn											
Upper	17	15	7.9	2.0-11.0	2.5	0.03	0.01-0.05	0.02	0.17	0.04-0.44	0.13
Lower	87[b]	10	35.1	12.0-58.0	15.5	0.04	0.01-0.06	0.02	0.84	0.08-2.26	0.69
Cookson	7.4	14	9.9	8.0-11.0	0.8	0.06	0.04-0.10	0.01	0.34	0.04-0.92	0.26
Fort Peck	185[c]	13	10.7	0.7-13.9	3.7	0.07	0.03-0.11	0.02	0.29	0.19-0.56	0.11
Sakakawea											
Upper	167[d]	24	13.2	3.0-27.0	5.5	0.05	0.01-0.10	0.03	0.46	0.06-0.98	0.27
Lower	252	21	22.6	3.0-40.0	11.6	0.06	0.02-0.10	0.02	0.47	0.15-0.86	0.21
Oahe											
Upper	165	21	16.4	3.7-44.4	6.8	0.05	0.02-0.09	0.02	0.76	0.29-1.55	0.37
Lower	344	20	38.6	6.7-50.3	13.9	0.06	0.02-0.08	0.01	0.52	0.18-0.92	0.16
Sharpe											
Upper	13	30	5.6	1.2-5.2	3.2	0.06	0.01-0.12	0.03	0.64	0.16-1.50	0.40
Lower	74	13	12.1	11.0-14.0	1.0	0.07	0.04-0.16	0.03	0.77	0.41-1.56	0.43
Francis Case											
Upper	65	13	5.6	4.9-6.4	0.6	0.02	0.01-0.03	0.01	0.46	0.18-0.78	0.17
Lower	171	13	27.1	10.8-38.4	11.8	0.06	0.03-0.08	0.01	2.78	1.01-4.54	1.02
Lewis and Clark	33	29	10.2	7.9-13.7	2.1	0.05	0.30-0.07	0.01	2.32	1.45-3.54	0.49
Nelson	8.5	17	7.3	4.0-10.0	1.8	0.03	0.01-0.05	0.01	-	-	-

[a]Tongue River data for mercury in sediments taken from a previous study (**Phillips 1979**).
[b]Transect located on side arm 1.5 km southeast of main channel.
[c]Transect located on side arm 20 km south of main channel.
[d]Transect located on side arm 10 km north of main channel.

Table 8. *Mercury concentrations in sediments (dry weight) and axial muscle of fish (wet weight) from various waters.*

Location	Mercury in sediment (μg/g)	Maximum Hg in fish (μg/g)	Species	Source
Antelope Reservoir (Oregon)	17.1[a]	1.79	Rainbow trout	Hill et al. (1975); Phillips and Buhler (1980)
Unspecified river (Manitoba)	0.01–109.0[b]	7.0	Not given	Langley (1973)
Lake Mývatn (Iceland)	0.01–0.04[b]	0.016	Arctic char	Olafsson (1979)
Hemlock Lake (Michigan)	0.02–1.25[b]	0.42	Rainbow trout	D'Itri et al. (1971)
American Falls Reservoir (Idaho)	0.21–0.95[b,c]	1.20	Rainbow trout	Kent and Johnson (1979)
Lake Powell (Arizona)	0.03[d]	0.76	Walleye	Potter et al. (1975)
Lohontan Reservoir (Nevada)	0.12–1.35[b]	2.72	White bass	Richins and Risser (1975)
Clay Lake (Ontario)	0.14–7.83[b]	16.0	Northern pike	Bligh (1970); Armstrong et al. (1972)
Section Four Lake (Michigan)	0.03–0.12[b]	0.45	Rainbow trout	D'Itri et al. (1971)
Lake Sangchris (Illinois)	0.05	0.30	Green sunfish	Anderson and Smith (1977)
Southern Indian Lake (Manitoba)	0.01[d]	0.51	Walleye	Bodaly and Hecky (1979)
Tongue River Reservoir (Montana)	0.04[d]	2.5	Northern pike	Phillips (1979)
Jocassee Reservoir (South Carolina)	0.04[a]	4.49[e]	Largemouth bass	Abernathy and Cumbie (1977)

[a]Only one sample taken.
[b]Range.
[c]Wet weight.
[d]Mean.
[e]Mean for size group of largest fish.

Table 9. *Correlations between concentrations of mercury and selenium, mercury and depth, and selenium and depth for surficial sediments in Missouri River Basin reservoirs.*

Reservoir location	No. of samples	Pearson correlation coefficient (r)[a]		
		Hg vs. Se	Hg vs. depth	Se vs. depth
Bighorn				
Upstream	15	0.15	0.84**	0.27
Downstream	9	−0.14	0.95**	−0.09
Cookson	14	−0.04	0.01	0.44*
Fort Peck	13	−0.07	0.78**	0.11
Sakakawea				
Upstream	24	−0.07	0.62**	0.11
Downstream	21	0.29	0.61**	0.42*
Oahe				
Upstream	21	0.78**	0.65**	0.45*
Downstream	20	0.28	0.69**	0.06
Sharpe				
Upstream	30	0.25	−0.02	−0.03
Downstream	13	0.25	−0.32	−0.41
Francis Case				
Upstream	13	0.46	0.38*	0.72**
Downstream	13	0.72**	0.76**	0.82**
Lewis and Clark	29	0.04	−0.23	0.23
Nelson	14	-	0.72**	-

[a]* = 5% significance level; ** = 1% significance level.

low only at locations where depth varied little along the entire transect. This tendency may result from the higher affinity of mercury for fine sediment particles that have a high surface area per unit volume; these particles tend to settle in the deepest portion of the basin (Thomas and Jaquet 1976). Moreover, sediments underlying deep water tend to be high in organic matter, which also binds mercury (Potter et al. 1975; Rust 1977).

For the most part, average mercury concentrations were not significantly different ($P \leq 0.05$) among locations. Exceptions included Nelson Reservoir and the upstream location in Lake Francis Case, where mercury concentrations were significantly lower than at several other locations; also, concentrations were significantly higher in the Big Dry Arm of Fort Peck than in upstream Bighorn Lake.

Selenium concentrations were more variable than mercury concentrations in sediments from the various locations. Mean concentrations ranged from 0.17 μg/g at the upstream end of Bighorn Lake to 2.78 μg/g at the downstream end of Lake Francis Case. Generally, selenium in sediment increased with downstream distance in the Missouri River watershed; however, the trend was statistically significant ($P \leq 0.05$) only for downstream Lake Francis Case and Lewis and Clark Lake. Most of the selenium in these downstream reservoirs apparently originates in the White River drainage (South Dakota). Soils in portions of Lyman and Gregory counties are naturally enriched with selenium, and agriculturally related selenium problems have occurred there for many years (Duane Murphy, South Dakota Department of Water and Natural Resources, personal communication). Mercury and selenium concentrations in sediments from most of the locations sampled were not correlated; however, the concentration of selenium, like that of mercury, was frequently correlated with water depth (Table 9).

No statistically significant trends related mercury or selenium concentrations to depth in the core samples. Mercury or selenium concentrations were higher in surficial than in deeper sediments in some locations, but the opposite was true in others (data not shown).

Mercury in Reservoir Walleyes

Walleyes from all 10 reservoirs showed a logarithmic pattern of mercury concentration relative to total length (Fig. 10; Table 10). In reservoirs where walleyes were taken from an upstream and downstream location there were no clear trends. For example, upstream walleyes accumulated mercury at a significantly faster rate than downstream walleyes in Bighorn Lake and Lake Oahe, whereas the opposite was true in Lakes Sharpe and Francis Case. We combined the data from both locations in each reservoir (Table 11) before making comparisons among reservoirs; combining the data increased the sample size and integrated conditions throughout an impoundment. We also ignored the differences in walleye growth rates among reservoirs because growth data were unavailable for the year of sample collection. Walleye growth in previous years was fastest in Tongue River Reservoir, Bighorn Lake,

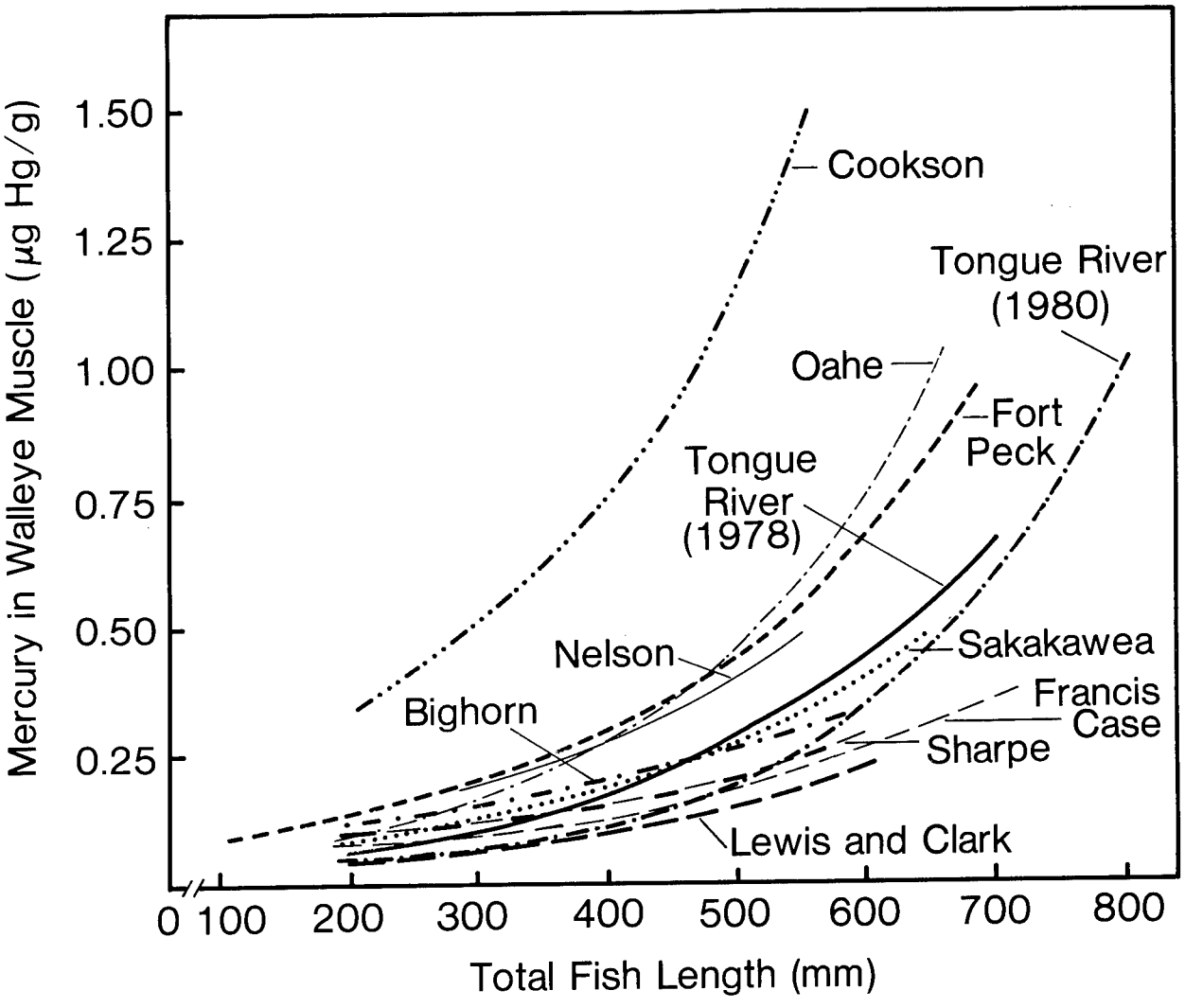

Fig. 10. Relations between concentration of total mercury (wet-weight basis) in axial muscle tissue and total length of walleyes collected from Cookson Reservoir, Lake Fort Peck, Bighorn Lake, Tongue River Reservoir, Lake Sakakawea, Lake Oahe, Lake Sharpe, and Lake Francis Case; and of saugers collected from Lewis and Clark Lake. Tongue River data portrayed collectively for 1978 through 1980 and for 1980 alone (the year in which all other reservoirs were sampled).

Table 10. *Relation between total length and mercury residues in walleyes or saugers*[a] *from 10 Missouri River Basin reservoirs.*

Reservoir and collection location	Sample size (n)	Total length range (mm)	Hg residue range ($\mu g/g$ wet weight)	Regression $\log_{10}$ (Hg) = a (length) $-b$	r^2
Cookson	47	190-555	0.11-1.60	$\log_{10}y = 0.0019x - 0.82$	0.35
Bighorn					
Upstream	50	190-653	0.08-2.15	$\log_{10}y = 0.0021x - 1.41$	0.73
Downstream	60	278-469	0.10-0.51	$\log_{10}y = 0.0014x - 1.08$	0.20
Fort Peck	97	216-688	0.10-1.28	$\log_{10}y = 0.0017x - 1.21$	0.53
Tongue					
1978	31	180-760	0.15-1.30	$\log_{10}y = 0.0018x - 1.45$	0.70
1979	9	395-745	0.27-1.55	$\log_{10}y = 0.0020x - 1.43$	0.86
1980	163	172-790	0.02-1.22	$\log_{10}y = 0.0022x - 1.76$	0.74
Sakakawea					
Upstream	99	227-622	0.04-0.51	$\log_{10}y = 0.0019x - 1.56$	0.56
Downstream	99	280-575	0.11-0.63	$\log_{10}y = 0.0013x - 1.16$	0.29
Oahe					
Upstream	53	240-528	0.09-0.51	$\log_{10}y = 0.0016x - 1.23$	0.50
Downstream	56	312-601	0.08-0.40	$\log_{10}y = 0.0019x - 1.57$	0.42
Sharpe					
Upstream	54	245-641	0.05-0.50	$\log_{10}y = 0.0020x - 1.79$	0.46
Downstream	51	230-564	0.05-0.42	$\log_{10}y = 0.0021x - 1.65$	0.58
Francis Case					
Upstream	60	322-515	0.05-0.26	$\log_{10}y = 0.0011x - 1.29$	0.10
Downstream	50	215-635	0.05-0.54	$\log_{10}y = 0.0016x - 1.29$	0.42
Lewis and Clark[a]	47	196-580	0.04-0.24	$\log_{10}y = 0.0015x - 1.59$	0.47
Nelson	28	272-540	0.08-0.52	$\log_{10}y = 0.0016x - 1.19$	0.46

[a]Saugers were substituted for walleyes in Lewis and Clark Lake.

and Lake Sakakawea, and slower and similar in the other reservoirs.

In general, mercury was accumulated at a faster rate in walleyes from reservoirs in the upriver portion of the drainage (Cookson, Bighorn, Nelson, Fort Peck, Tongue) than in walleyes from downstream reservoirs (Fig. 10). The rate was fastest in Cookson Reservoir ($P \leq 0.01$) and faster in Bighorn, Fort Peck, and Nelson reservoirs ($P \leq 0.01$) than in the reservoirs from Lake Sakakawea downstream (Table 11). Mercury in sediment was unrelated to mercury content of fish.

For the main stem Missouri reservoirs, the rate of mercury uptake relative to walleye length was sequentially related to distance downstream—i.e., the farther downstream the reservoir, the slower the rate of mercury accumulation. Growth rates of walleyes were similar in upstream and downstream reaches. Mercury concentration in fish (relative to total fish length) was positively correlated ($P \leq 0.01$) with the ratio of maximum daily inflow to average daily inflow and negatively correlated ($P \leq 0.01$) with the percentage of inflow water that had previously been impounded (Table 12). Thus upstream reservoirs with less controlled inflows (and thus more severe flooding) were more likely to have fish with higher mercury concentrations.

Uthe et al. (1973) and Gummer (1980) have shown that erosion and scouring that occur during high flows mobilize mercury present in surficial river sediments, resulting in a pulse of mercury movement. Miller (1977) and Park et al. (1980) showed similar evidence for a methylmercury pulse. Our results concur with the concept that upstream flooding is an important factor in the accumulation of mercury by reservoir fish.

Table 11. *Regressions of mercury concentration (wet weight) in axial muscle tissue against total length for walleyes collected from 10 Missouri River Basin reservoirs. Data from different sampling stations within a reservoir were combined.*

Location	Sample size (n)	Regression[a] Log_{10} (Hg) = a (length) $-b$	Predicted Hg concentration (μg/g) in walleyes of different lengths		
			400 mm	500 mm	600 mm
Cookson	47	$\log_{10}y = 0.0019x-0.82$	0.87	1.35	2.08
Bighorn	110	$\log_{10}y = 0.0020x-1.33$	0.30	0.42	0.71
Fort Peck	97	$\log_{10}y = 0.0017x-1.21$	0.30	0.44	0.65
Nelson	28	$\log_{10}y = 0.0016x-1.19$	0.28	0.41	0.59
Tongue (1979)	9	$\log_{10}y = 0.0020x-1.43$	0.23	0.37	0.59
Tongue (1978)	31	$\log_{10}y = 0.0018x-1.45$	0.19	0.28	0.43
Sakakawea	198	$\log_{10}y = 0.0016x-1.38$	0.18	0.26	0.38
Tongue (1980)	163	$\log_{10}y = 0.0022x-1.76$	0.13	0.22	0.36
Oahe	109	$\log_{10}y = 0.0010x-1.07$	0.21	0.27	0.34
Sharpe	105	$\log_{10}y = 0.0018x-1.62$	0.13	0.19	0.29
Francis Case	110	$\log_{10}y = 0.0011x-1.23$	0.16	0.21	0.27
Lewis and Clark[b]	47	$\log_{10}y = 0.0015x-1.59$	0.10	0.14	0.20

[a]Mercury data for upstream and downstream sampling locations were combined for the reservoirs in which both locations were sampled.
[b]Saugers were substituted for walleyes in Lewis and Clark Lake.

Table 12. *Pearson correlation coefficients between the average mercury concentration (wet weight) in walleyes 500 mm long from each of 10 reservoirs in the Upper Missouri River (computed from regressions) and various physical, chemical, and biological characteristics.*

Characteristics	Comparisons (no.)	Pearson correlation coefficient	P[a]
General characteristics			
Mean mercury in sediments (μg/g)	16	0.13	0.31
Mean selenium in sediments (μg/g)	13	−0.26	0.20
Time since dam closure (years)	20	−0.38	0.06
Outflow height from bottom (m)	10	−0.39	0.15
Mean depth of sediment transect (m)	14	−0.10	0.37
Maximum depth of sediment transect (m)	15	−0.16	0.30
Reservoir surface area (km^2)	20	−0.30	0.11
Reservoir volume ($10^9 \times m^3$)	20	−0.27	0.14
Maximum depth at station sampled (m)	19	0.32	0.10
Daily net primary productivity (g C/m^2)	9	0.50	0.08
Ratio of maximum inflow to average inflow	18	0.97	0.00**
Steady state total phosphorus (μg/L)	14	−0.12	0.36
Percent inflow water previously impounded	16	−0.59	0.00**
Phytoplankton standing crop (mm^3/L)	10	0.34	0.16
Chlorophyll a (mg/m^3)	14	−0.07	0.41

Table 12. *Continued.*

Characteristics	Comparisons (no.)	Pearson correlation coefficient	P^a
Annual sediment deposition volume ($m^3/km^2 \times 10^4$)	11	−0.08	0.40
Conductivity (μmhos/cm)			
Minimum surface	13	0.82	0.00**
Maximum surface	13	0.76	0.00**
Mean surface	13	0.85	0.00**
Minimum bottom	11	0.69	0.01**
Maximum bottom	11	0.55	0.05*
Mean bottom	11	0.67	0.02**
pH			
Minimum surface	18	0.75	0.00**
Maximum surface	18	0.51	0.02*
Mean surface	18	0.71	0.00**
Minimum bottom	18	0.74	0.00**
Maximum bottom	18	0.59	0.01**
Mean bottom	18	0.70	0.00**
Dissolved oxygen (mg/L)			
Minimum surface	19	−0.53	0.01**
Maximum surface	19	0.04	0.49
Mean surface	19	−0.27	0.14
Minimum bottom	19	−0.05	0.42
Maximum bottom	19	0.15	0.27
Mean bottom	19	0.02	0.48
Water temperature (°C)			
Minimum surface	19	−0.13	0.31
Maximum surface	19	−0.06	0.41
Mean surface	19	−0.15	0.28
Minimum bottom	19	0.03	0.44
Maximum bottom	19	0.14	0.29
Mean bottom	19	0.11	0.33
Turbidity (JTU)			
Minimum surface	15	0.25	0.19
Maximum surface	15	0.68	0.00**
Mean surface	15	0.64	0.01**
Minimum bottom	15	0.60	0.04*
Maximum bottom	15	0.46	0.11
Mean bottom	15	0.56	0.06
Nonfilterable solids (mg/L)			
Minimum surface	11	0.82	0.00**
Maximum surface	11	0.84	0.00**
Mean surface	11	0.85	0.00**
Turnover rate (times/month)			
Minimum	20	−0.30	0.12
Maximum	20	−0.34	0.08
Mean	20	−0.34	0.09
Total surface dissolved solids (mg/L)			
Minimum	13	0.76	0.00**
Maximum	13	0.74	0.00**
Mean	13	0.81	0.00**

[a]* = 95% significance level; ** = 99% significance level.

Characteristics of Missouri River Basin reservoirs that were consistently correlated with mercury uptake by fish included pH, conductivity, total dissolved solids, and nonfilterable solids (Table 12). It was shown by deFreitas et al. (1977) that an increase in pH from 8.0 to 8.5 accelerated monomethylmercury accumulation, as did increasing water hardness. Our findings are consistent with their results. Others, however, have noted a negative relation between methylmercury uptake and both pH and water hardness (Drummond et al. 1974; Rogers and Beamish 1983; Wren and MacCrimmon 1983). Such inconsistency serves to point out the complex interactions that undoubtedly occur between variables that influence methylation and bioavailability of mercury. Clearly, no single rule of thumb can confidently be applied to all bodies of water.

Inasmuch as mercury associates with the particulate material in water, and organic particulates provide substrates for bacterial growth, turbidity composed of organic particulate material may facilitate methylation of mercury by promoting bacterial growth. Furutani and Rudd (1980) found that the methylating activity of both water and sediment floc was substantially increased by the addition of organic nutrients that could be used by bacteria, and that formation of methylmercury was strongly correlated with microbial activity. Rudd and Turner (1983) showed that suspension of lake sediments in the water column decreased inorganic mercury accumulation by fish; however, only inorganic sediments were suspended. Flooding of terrestrial soils probably increases the loading of terrestrially mediated methylmercury. More research on methylation of mercury is clearly needed.

Walleyes from Cookson Reservoir contained significantly more mercury ($P \leq 0.01$) relative to length than did walleyes from any other reservoir in our study (Table 11). This high mercury concentration may be related to the relative youth of the reservoir (6 years in 1981). Reservoir age seemed negatively related to mercury content of fish, but the significance was marginal ($P = 0.06$). Abernathy and Cumbie (1977), Bodaly and Hecky (1979), and Cox et al. (1979) all noticed that resident fish tended to have unusually high mercury concentrations for the first several years after reservoirs were impounded. They attributed this high concentration to the leaching of mercury from the soil after the initial inundation. In all reservoirs, mercury concentrations declined in fish after several years, apparently as a result of the sequestering of mercury by humic materials.

Selenium in Reservoir Walleyes

Selenium residues in tissues of walleyes from Bighorn Lake, Lake Sharpe, and Lake Fort Peck were not correlated with fish length nor with mercury in tissue (Table 13). This lack of correlation differs from findings for marine fish, in which selenium and mercury occur at a relatively fixed molar ratio (Nakagawasai et al. 1976; Friedman et al. 1978); however, our findings agree with those of Cappon and Smith (1981) for freshwater fish. Speyer (1980), comparing northern pike from two Quebec lakes, showed that fish from one lake contained high selenium and low mercury concentrations, whereas the opposite was true for fish from the other lake. However, concentrations of mercury and selenium also differed in sediments from the two lakes and corresponded to the concentrations of these elements in the fish. Ratios in fish may thus simply reflect the relative concentrations of mercury and selenium in the environment.

Our analyses of residues in tissues do not support the belief that the presence of selenium affects the mercury content of reservoir fish. The bioaccumulative tendencies of methylmercury are partly ascribed to its ability to readily exchange positions between different sulfhydryl binding sites at the surfaces of membranes. One would consequently infer that selenium would interfere with mercury uptake since mercury has a stronger affinity for selenium than for sulfur (Carty and

Table 13. *Pearson correlations (r) between selenium concentration (wet weight) in axial muscle tissue and both total length and mercury concentration (wet weight) in axial muscle tissue of walleyes from three Missouri River Basin reservoirs (P > 0.05 for all r's).*

Reservoir	n	Total length (r)	Mercury in muscle (r)
Bighorn Lake	50	0.10	−0.13
Lake Sharpe	54	0.12	0.11
Fort Peck Lake	97	−0.02	−0.14

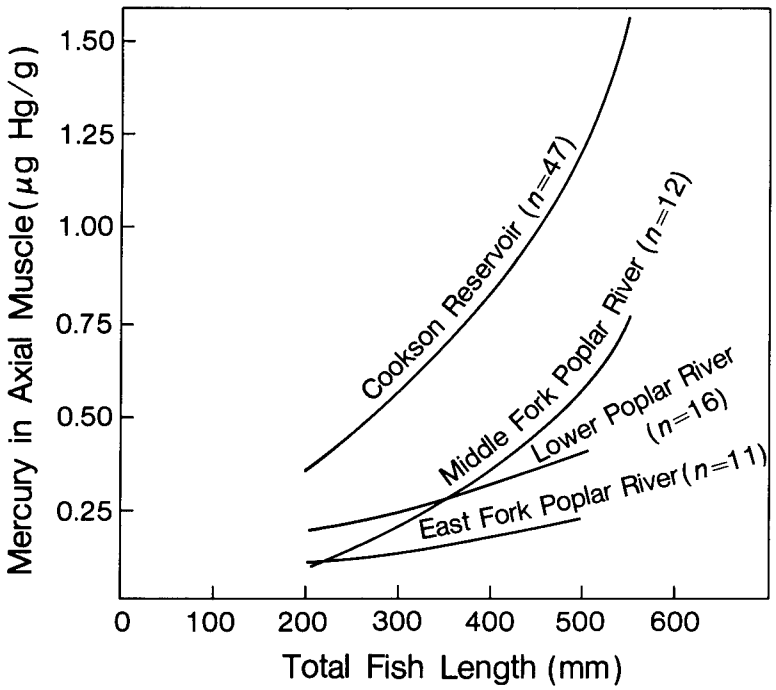

Fig. 11. Relations between concentration of total mercury (wet weight) in axial muscle and total length of walleyes collected from Cookson Reservoir and the East Fork, Middle Fork, and Lower Poplar rivers.

Malone 1979); however, our results do not support this.

Reservoir Fish versus River Fish

Walleyes from all three branches (East Fork, Middle Fork, Lower) of the Poplar River, downstream from Cookson Reservoir contained significantly less mercury relative to length ($P < 0.01$) than did walleyes from Cookson Reservoir (Fig. 11). Phillips (1979) noticed a similar relation between crappies in Tongue River and Tongue River Reservoir, as did Walter et al. (1974) for fishes of Oahe Reservoir and its tailwaters. Walleyes in the East Fork (the impounded branch) also contained significantly less mercury in relation to length than did walleyes from the Lower Poplar ($P = 0.03$). The mercury content of East Fork walleyes did not differ from that of Middle Fork walleyes ($P = 0.15$) when all of the data were included; however, the difference became strongly significant ($P = 0.01$) when one East Fork fish (which contained the highest mercury concentration) was excluded. Alternatively, the mean mercury concentration in 71 common carp collected from Tongue River several miles upstream from Tongue River Reservoir did not differ (t-test) from that in 56 common carp of similar size from the reservoir. Concentrations of mercury were not as well correlated with length in common carp as in other fish species that we sampled, possibly due to greater variation in growth. The highest mercury concentrations in common carp were similar to the highest concentrations in saugers and walleyes. Conditions for mercury accumulation were seemingly more favorable in Cookson and Tongue River reservoirs than in the rivers downstream.

Mercury Uptake by Northern Pike

Northern pike taken from Tongue River Reservoir in 1978–1981 showed a logarithmic pattern of mercury uptake (relative to length) during each of the 4 years of sampling (Fig. 12). This pattern was noted for all species and locations sampled during this study, presumably because fish-length intervals spanning consecutive age-groups become increasingly shorter as fish increase in age (i.e., growth curves tend to plateau with age). The positive correlations between fish size and mercury concentrations in tissue (Johnels et al. 1967; Scott 1974; Olsson 1976) result from the slow elimina-

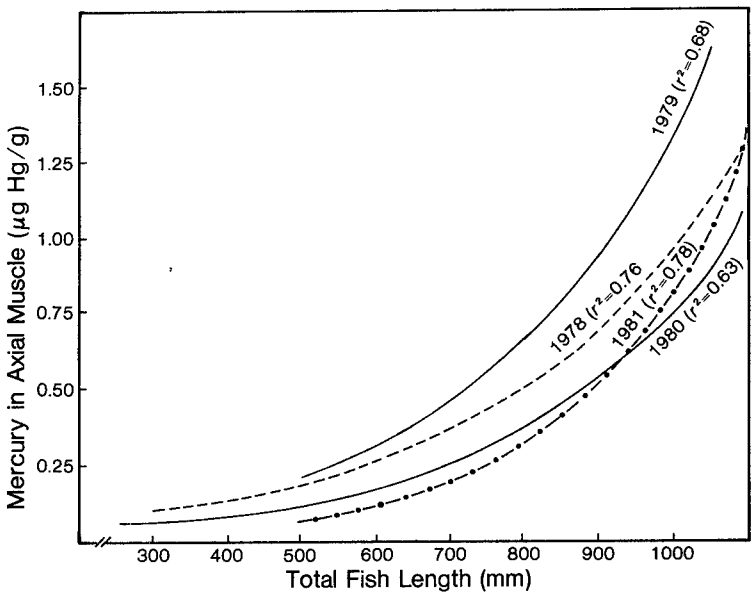

Fig. 12. Relations between concentration of total mercury (wet weight) in axial muscle and total length of northern pike collected from Tongue River Reservoir in spring, 1978–1981. (Sex ratios were similar in the different years.)

tion of MeHg. The biological half-life of MeHg in fish is reportedly as much as 7 years (Knight 1982).

In Tongue River Reservoir mercury was accumulated at a faster rate in northern pike than in other fishes, suggesting that the pike either were exposed to more MeHg or accumulated it more efficiently than other species. Olsson (1976) found a higher correlation between mercury concentration and fish length than between mercury concentration and fish age and concluded that metabolic rate was more important than exposure time in determining mercury residues in fish tissue. However, deFreitas et al. (1977) concluded that growth dilution by faster growing species resulted in lower mercury concentrations in tissue; their conclusion is consistent with our observation that mercury concentrations in northern pike of a given length from Tongue River Reservoir were lower in the faster growing females than in males of the same age (Phillips et al. 1980). We conclude that the northern pike were exposed to more mercury than the other species because they ate larger food organisms, which contained more mercury.

Northern pike collected in spring 1979 contained significantly higher mercury concentrations relative to length than did those taken during any of the other years of sampling (Fig. 12). The elevated mercury concentration coincided with a 100-year flood in spring 1978 (Fig. 13). Bodaly and Hecky (1979) found higher mercury concentrations in northern pike from Southern Indian Lake, Manitoba, after a flood than before it, and Uthe et al. (1973) attributed the increased rate of mercury uptake during July by caged rainbow trout in the south Saskatchewan River to the fresh deposition of mercury-laden sediments mobilized during spring floods. Methylmercury is produced in terrestrial soils (Rogers 1977) and tends to accumulate in surface soil horizons (Andersson 1979). Inundation desorbs MeHg trapped in soils and facilitates its transport (Ottawa River Project Group 1979).

Flooding may also stimulate MeHg production in the water column. Jernelöv and Åsell (1975) showed that the agitation of lake sediments spiked with inorganic mercury greatly increased methylmercury production. These experiments simulated the disturbance that occurs at the sediment-water and soil-water interfaces during flooding. Methylmercury concentrations in the water of Wabigoon River, Manitoba, increased with distance downstream after the spring flood subsided but total mercury concentrations did not. Jackson and Woychuk (1980) interpreted this downstream change in MeHg concentration as evidence that mercury associated with suspended particulate matter was being methylated in the water column downstream. Furutani and Rudd

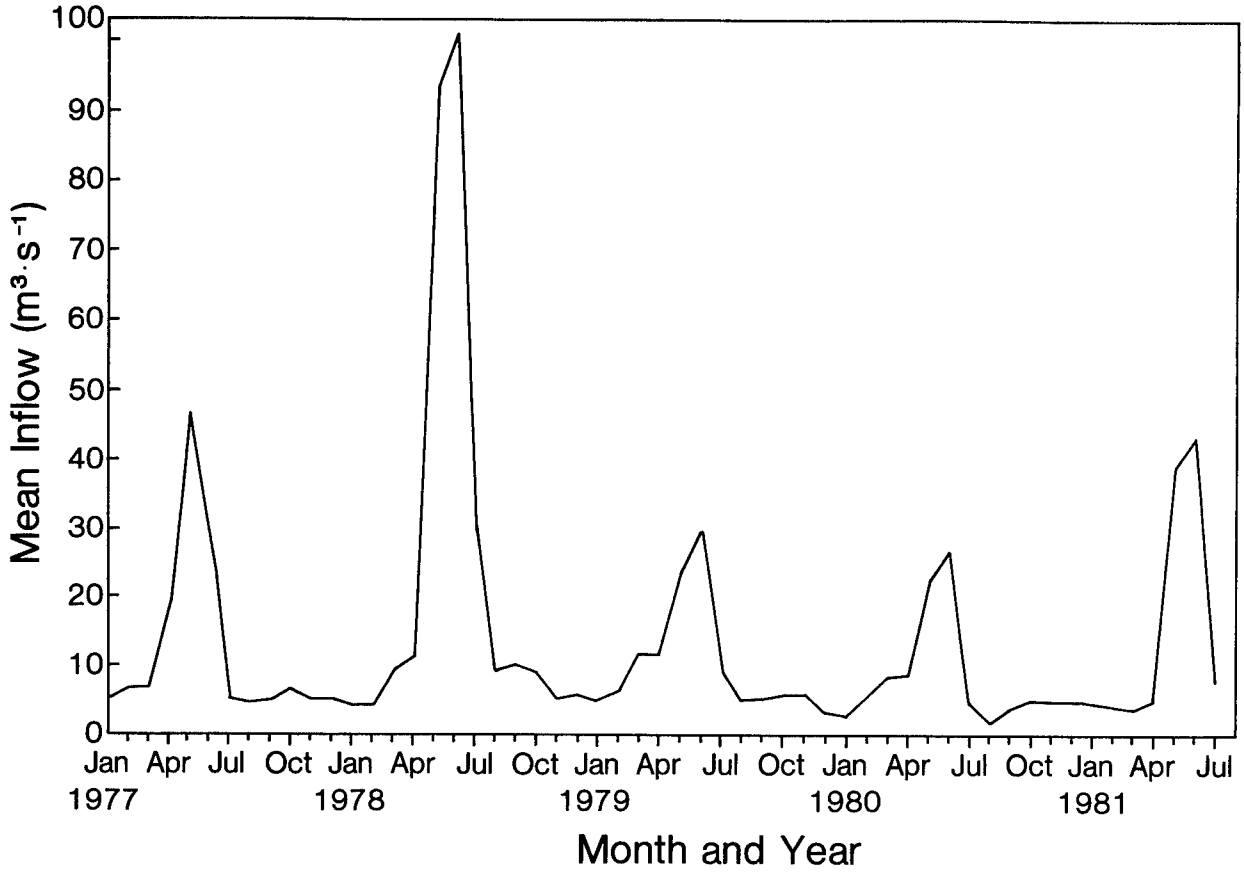

Fig. 13. Water flow rates in the Tongue River immediately upstream from Tongue River Reservoir, 1978–1981.

(1980) showed that the methylation rate was greatest during spring runoff due to the concomitant increase in nutrients in the water. The present study and others thus indicate that flooding is an important aspect of mercury mobilization and methylation.

Mercury Uptake by Individual Northern Pike

Mercury concentrations in nine northern pike biopsied in both 1978 and 1979 increased over the interval (Fig. 14); conversely, the concentration decreased in four of five northern pike biopsied in 1979 and 1980 and in two of three sampled in 1979 and again in 1981. Concentration remained relatively stable between 1980 and 1981; observed changes (in μg/g) were 1.60 to 1.71, 0.89 to 0.64, and 0.34 to 0.36. Lockhart et al. (1972), who analyzed individual northern pike transferred from a mercury-contaminated lake to a pristine lake, reported that the mercury concentration in white muscle of the fish decreased to 27% of the initial concentration after 1 year; however, most of the change could be accounted for by dilution resulting from growth. Growth dilution could also account for the decrease in mercury concentration between 1979 and 1981 in northern pike from Tongue River Reservoir.

As judged by approximate growth curves for male and female northern pike, the growth of northern pike in Tongue River Reservoir was rapid and females grew faster than males (Fig. 15). Length estimates for the first two age-groups were determined from recaptures of marked fish of known age, and lengths of older age-groups were estimated from mark-and-recapture data by overlapping similar size categories for fish that had been captured and measured at least twice (in different years).

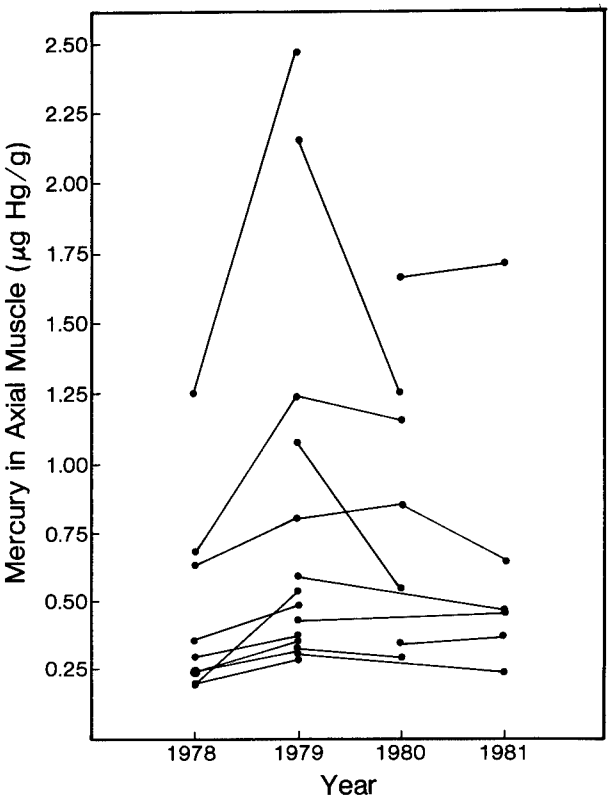

Fig. 14. Mercury concentration (wet weight) in individual northern pike in the Tongue River Reservoir, biopsied and analyzed for mercury (wet weight) at least twice between spring 1978 and spring 1981.

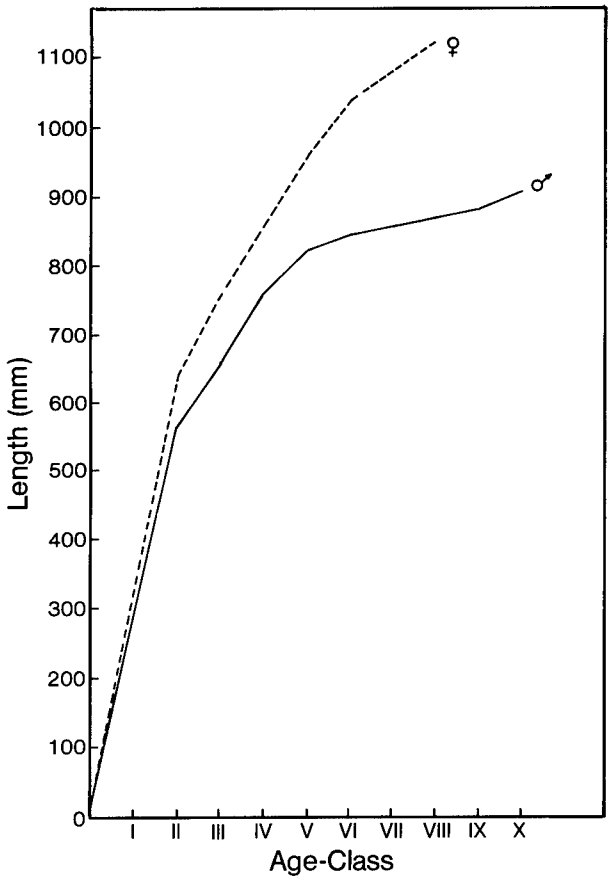

Fig. 15. Interpolated growth curves for male and female northern pike in the Tongue River Reservoir (see text for discussion).

Mass Balance Budget

Our mass balance budget indicated that the Tongue River was the primary source of mercury to Tongue River Reservoir and that most of the mercury was from nonpoint sources (Table 14). Kudo (1977*a*) noted a similar situation in the Ottawa River in Canada. Natural weathering of rocks and soil movement supply most of the mercury to relatively uncontaminated systems. The proportion of mercury entering from the atmosphere should be much greater in seas or large lakes such as the Great Lakes (Brzezinska and Garbalewski 1980) than in a reservoir system because atmospheric input becomes more significant as the ratio of surface area to volume increases and retention time increases.

Overburden and interburden at North Decker and East Decker mine sites contained $\leq$0.001–0.62 μg/g of mercury (Table 15) but the settling-pond effluents of coal mines accounted for only 1% of the mercury entering the reservoir from point sources. The largest point source was the municipal sewage treatment plant at Sheridan, Wyoming, which accounted for almost 9% of the mercury entering the reservoir; average concentration in the effluent was 0.16 μg/L. Similarly, Chen et al. (1974) reported that secondary effluent from a sewage treatment plant in California contained 0.16 μg/L. In a recent EPA study of 40 representative sewage treatment plants nationwide, detect-

Table 14. *Estimated mercury fluxes to and from Tongue River Reservoir, April 1980–March 1981.*

Flux	Mercury input rate (g/year)	Percent of total input to river or reservoir
Inputs to Tongue River		
Sewage treatment plant	385	9.6
Bighorn Mine		
Upper pond	12	0.3
Lower pond	6	0.1
Other sources[a]	3,620	90.0
Total	4,023	100.0
Inputs to Tongue River Reservoir		
East Decker Mine		
South pond	18	0.4
North pond	11	0.2
Tongue River inflow	4,023	93.4
Intermittent streams	56	1.3
Groundwater	1	0.02
Precipitation	192	4.5
Dry deposition	5	0.1
Total (from monitored sources)	4,306	100.0
Output from reservoir		
Tongue River outflow	5,435	126.2
Net loss	1,129	

[a]Because no other significant point sources are known, we believe this category consists primarily of nonpoint sources, including natural weathering and erosion.

Table 15. *Mercury concentrations in overburden and interburden at coal mining sites, as reported by Hittman Associates, Inc. (1981c).*[a]

Location and sample material	No. of samples	Mercury concentration (μg/g)		
		Mean	SD	Range
North Decker Mine area				
Dietz 1 Overburden	34	0.04[b]	0.03	≤0.001-0.11
Clinker	22	<0.01[b]	0.01	≤0.001-0.03
Dietz 1-2 Interburden	52	0.06[b]	0.04	≤0.001-0.02
Dietz 3 Interburden	188	0.05[b]	0.05	0.001-0.51
Overburden	120	0.07	0.08	0.001-0.62
East Decker Mine area				
Overburden	196	0.0024	0.004	<0.001-0.02
Interburden				
Total mercury	1	0.04	—	—
Extractable mercury	1	<0.001	—	—

[a]Data of Hittman Associates, Inc., North Decker Mine Plan, East Decker and North Extension Mines, Draft Environmental Impact Statement, Volume 2.
[b]Weighted mean values.

able mercury in effluents ranged from 0.20 to 1.20 μg/L and averaged 0.55 μg/L. (Burns and Roe 1982). Inasmuch as the mercury content of raw sewage averages 2 μg/L (Matheson 1979), environmental inputs near metropolitan areas can be significant. The National Academy of Sciences (1978) reported that nearly 19% of the anthropogenic inputs of mercury to water are from sewage.

The reservoir at full pool contained 1.47×10^3g of mercury in 1981, which is about 33% of the annual mercury inflow. Such a ratio indicates the importance of the river effect on this reservoir and illustrates that mercury moves rapidly through Tongue River Reservoir.

Our estimates suggested that more mercury left than entered Tongue River Reservoir from April 1980 to March 1981. Calculations for two additional periods, October 1978 to September 1979 (Phillips 1979) and October 1980 to September 1981 (unpublished data), yielded the following exchange (in grams Hg per year): for 1978-1979 (a period including the 100-year flood that resulted in fish having high concentrations of mercury), an inflow of 7.5×10^3 and an outflow of 7.1×10^3; and for 1980-1981 an inflow of 5.2×10^3 and an outflow of 6.2×10^3. These estimates suggest considerable variation between years. During the present study, spring runoff was minor and scouring of mercury from terrestrial soils and river sediments was presumably small. This evidence further supports our contention that nonpoint sources accounted for most of the mercury in the reservoir. Additionally, light snow cover in 1980 may have resulted in more mercury being deposited on and bonded to soil, rather than deposited on snow and washed into streams with meltwater.

Transport by streambed load was probably responsible for much of the remaining mercury entering the reservoir, since the reservoir acts as a catch basin for sediments. Townsend and Kudo (1977) estimated that about 1% of the mercury transported during quiescent conditions in the Ottawa River was by sediment movement. However, streambed movement is greatest during the spring flood and could contribute substantially to mercury movement—particularly since most mercury in freshwater systems is bound to sediments (Kudo 1977*b*; Jernelöv 1980).

Peak inputs may have been missed by our sampling. Samples of storm water (Hittman Associates, Inc.1981*b*) indicated a 1-day period in May 1980 or 1981 of elevated mercury concentrations (0.3-0.4 μg/L) in Tongue River—substantially above our range of 0.01-0.03 μg/L. Pulse inputs are also possible from coal mines as layers of differing mercury concentrations are disturbed and exposed to leaching.

Table 16. *Number and size range of walleyes and white crappies from Tongue River Reservoir collected during four sampling periods in 1980.*

Sampling dates	Walleyes			Crappies		
	n	Total length (mm)	Weight (*g*)	*n*	Total length (mm)	Weight (*g*)
April 16-May 9	34	436-700	820-3,600	78	163-382	60-780
June 18-July 10	18	223-675	90-2,600	46	190-258	90-210
August 7-14	59	172-655	50-2,620	65	138-288	30-340
September 30-October 8	52	190-790	80-6,210	57	154-296	40-385

We did not incorporate volatilization of deposition of elemental mercury (Hg°) at the air-water interface into this budget because we lacked the needed information. A review of pertinent literature indicated that mercury is probably lost from alkaline waters by volatilization (Fagerström and Jernelöv 1972). Matheson (1979) noted that surface waters are not strong sinks for elemental mercury; however, oxidation to Hg^{2+} can occur rapidly in the water column. Release from a lake to the atmosphere may remove much of the mercury deposited by rainfall (Jernelöv 1980). The need for studies at the air-water interface is obvious.

Our mass balance calculations should be interpreted cautiously because data were collected over a relatively short time, and not all point and nonpoint sources were monitored. In addition, uncertainties in our flux estimates are conceivably large. Although we documented few effects from present mining, future impacts of intense mining in the Tongue River drainage could cumulatively become significant.

Accumulation of Dietary Mercury by Fish

A total of 163 walleyes (172-790 mm long) and 247 white crappies (138-382 mm long) were collected from Tongue River Reservoir in 1980 (Table 16). Size distribution varied among sampling periods. In walleyes, large fish predominated in the catch in April and small fish in June and August, and sizes were rather evenly distributed in October; in crappies, size distribution was more even, although large crappies were rarely caught in June and yearlings did not appear in the catch until August.

Mercury in Walleyes and White Crappies

Mercury concentrations in axial muscle tissue of walleyes and white crappies increased exponentially with increasing fish length (Figs. 16 and 17), as previously reported by Phillips (1978, 1979). In other waters, relations of fish size to mercury concentration have been reported to be positive for a variety of species (Bache et al. 1971; Scott and Armstrong 1972; Potter et al. 1975; Richins and Risser 1975; Benson et al. 1976; Cox et al. 1979; Hildebrand et al. 1980) and were characterized as being exponential by Scott (1974). Increases in mercury concentration with size and age appear to be nearly universal among long-lived piscivorous fishes.

Mercury concentrations ranged from 0.02 to 1.22 μg/g in walleyes and from 0.02 to 0.53 μg/g in white crappies; the concentration in 2 of the 163 walleyes, but none of the 248 white crappies, exceeded 1.0 μg/g wet weight. Mercury concentrations were higher in walleyes than in white crappies of similar age (Fig. 18); these values were estimated by using the mean length for each age-group in the appropriate regression equation for mercury concentration against length. The magnitude of the difference between the two species became more pronounced with age, and appeared to stem from dietary changes that resulted in differential rates of mercury consumption; details of this relation are discussed later.

Mercury concentrations in muscle tissue were similar to whole-body values. In homogenized whole-body tissues of 8 walleyes and 18 white crappies, average mercury concentrations were 0.08 and 0.10 μg/g, respectively. Regression equations of mercury concentration in muscle against total

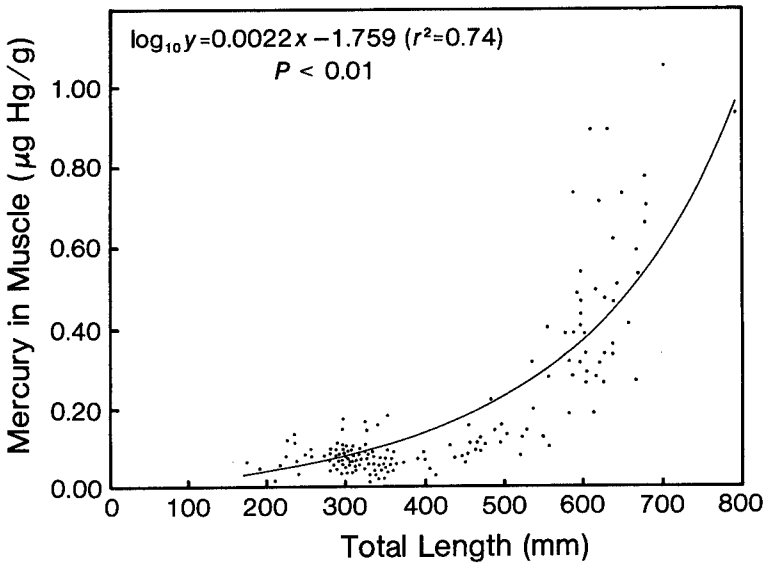

Fig. 16. Relation between concentration (wet weight) of total mercury in axial muscle and total length of 163 walleyes collected from Tongue River Reservoir in 1980. Fish from all four sampling periods are included.

length for these species (Figs. 16 and 17) predicted that average concentrations in fish of equivalent mean length (walleyes, 316 mm; white crappies, 199 mm) should be 0.09 and 0.11 μg/g. These observations, combined with similar findings (Miettinen et al. 1970; Lockhart et al. 1972; McKim et al. 1976; Phillips 1978; Ribeyre and Boudou 1980), justify the use of mercury concentrations in muscle to estimate whole-body mercury concentrations.

Concentrations of MeHg were generally at or below detection limits because mercury concentrations were low in the few fish analyzed for MeHg. Collection of larger fish for MeHg analysis probably would have yielded different results. For the one fish in which MeHg concentration clearly exceeded our detection limit of 0.10 μg MeHg/g, MeHg made up 73% of all mercury present. This percentage is lower than most reported mean values (Knight 1982); however, no conclusions can

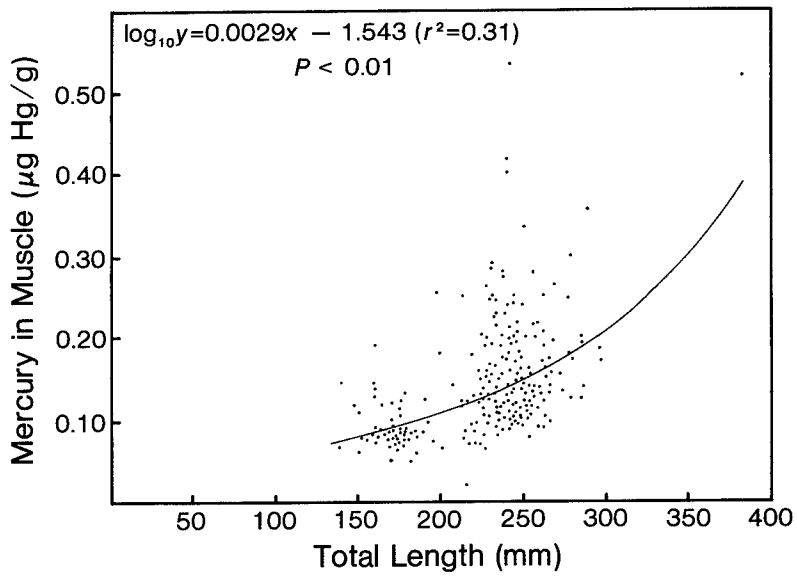

Fig. 17. Relation between concentration (wet weight) of total mercury in axial muscle and total length of 248 white crappies collected from Tongue River Reservoir during 1980. Fish from all four sampling periods are included.

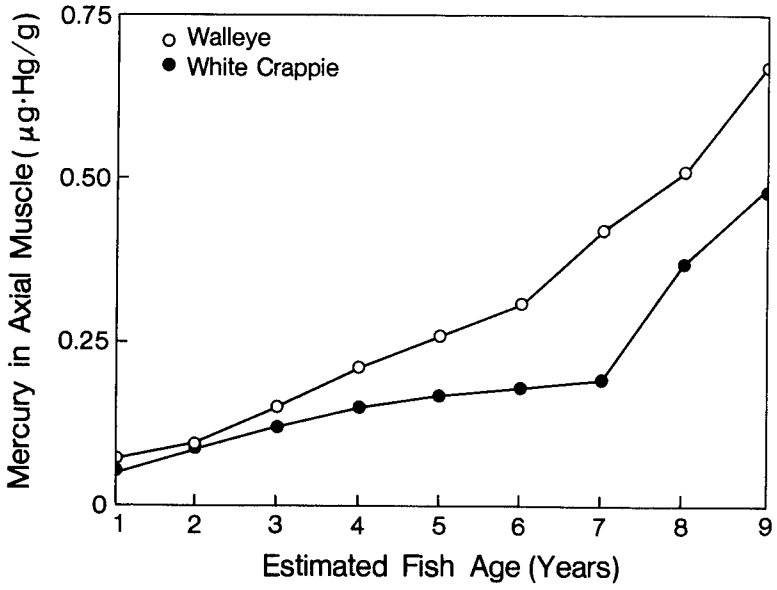

Fig. 18. Predicted mean total mercury concentrations (wet weight) in walleyes and white crappies of the same ages from Tongue River Reservoir, 1980. See text for derivation of mercury values. Age-group estimates are based on studies of walleyes by Riggs (1978) and of white crappies by Elser et al. (1977).

be drawn from a single sample because variability about the mean can be 20% or more. Consequently, in all calculations we used values taken from the literature for the percentage of total mercury present as MeHg.

Food of Walleyes and White Crappies

All length-classes of walleyes in Tongue River Reservoir were predominantly piscivorous; fish constituted about 80–100% of the food volumes in stomachs and occurred in all but two of the stomachs sampled (Knight 1982). Invertebrates were found only in the stomachs of walleyes shorter than 350 mm and were important only in walleyes shorter than 250 mm, in which they occurred in 54% of the stomachs and composed 20% of the total food volume.

Average and maximum size of fish eaten increased with walleye size (Fig.19); however, the

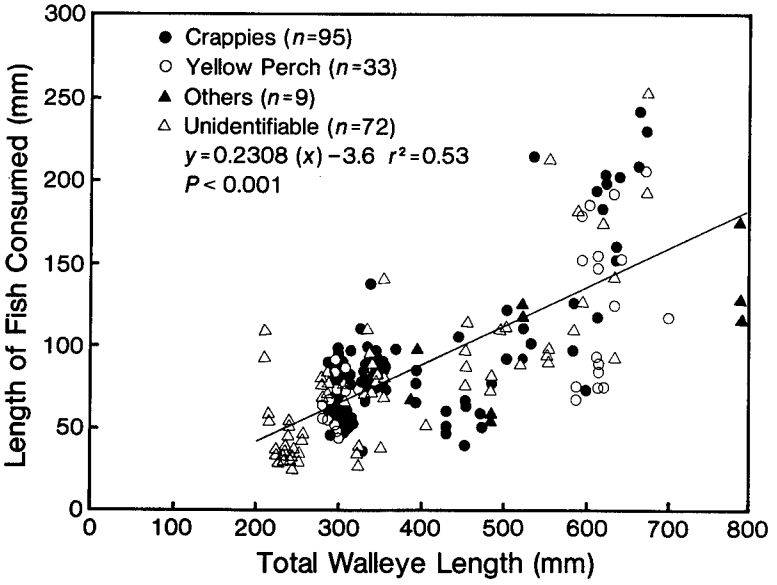

Fig. 19. Relation of estimated total lengths of fish eaten by walleyes to walleye total length.

minimum size of fish eaten changed little, because walleyes ate young-of-the-year fish when they were available. Differences in length of forage fish eaten by walleyes of different length-classes were greatest in April, when no young-of-the-year fish were available. Parsons (1971) and Forney (1974) also found that walleyes selected for young-of-the-fish, and that larger walleyes ate larger forage fish when the young fish were unavailable. Stomach-content volumes and the frequency of empty stomachs (average, 15%) also increased with walleye length, suggesting that older walleyes fed less frequently but ate larger meals. Individual stomach-content volumes varied greatly; however, sample sizes were large enough to compensate, as evidenced by the relatively small standard errors.

Crappies were the principal food of walleyes throughout the year (Fig. 20), accounting for 36-76% of the food volume and occurring in 43% of the stomachs examined (Knight 1982). Yellow perch were also eaten regularly but in smaller amounts (0-18% of the volume). Most of the unidentified fish in stomachs appeared to be one of these two species.

Changes in the diet of walleyes, with both season and size, seemed related to food abundance and food size (Fig. 21). In spring, young walleyes ate invertebrates (primarily chironomid larvae and pupae) and whatever small forage fish were available, and larger walleyes ate larger forage fish. During July, young-of-the-year crappies became available and walleyes of all sizes began feeding heavily on them. Young-of-the-year crappies decreased in importance as a forage item in October, perhaps as the combined result of a decline in numbers and growth beyond the optimal forage size for walleyes. Walleyes then diversified their diets and the larger individuals began feeding on larger forage fish.

Walleyes apparently fed opportunistically on white crappies, the most abundant forage fish species in Tongue River Reservoir (Elser et al. 1977). Forney (1974) reported that seasonal dietary patterns of walleyes in Oneida Lake, New York, were related to changes in the availability and size of the predominant forage fish. The variety of fish species eaten by walleyes in different water bodies provides further evidence that walleyes are opportunistic feeders (Priegel 1963; Wagner 1972; Swenson 1977).

In waters where yellow perch are the predominant forage species, variations in year-class

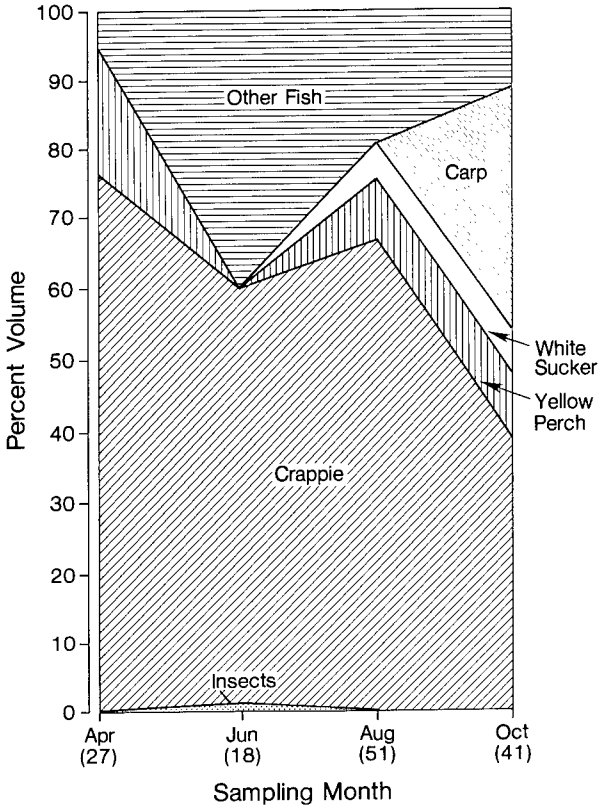

Fig. 20. Seasonal composition of stomach contents of walleyes from Tongue River Reservoir, 1980. Sample size is shown in parentheses below month.

strengths of walleyes and yellow perch are often synchronous (Forney 1974, 1977; Swenson and Smith 1976; Swenson 1977). High prey densities provided by a strong year-class of yellow perch seemed to decrease the incidence of cannibalism by walleyes, further strengthening the walleye year-class (Chevalier 1973). We observed no cannibalism in walleyes of Tongue River Reservoir.

Fish were also an important food of white crappies in Tongue River Reservoir, occurring in 46% of the stomachs and contributing 78% of the food volume (Knight 1982); the stomachs of crappies longer than 255 mm contained the highest percentages of fish (by volume). Cannibalism on young-of-the-year white crappies was prevalent among all sizes of crappies sampled. Although cannibalism by white crappies has occasionally been reported

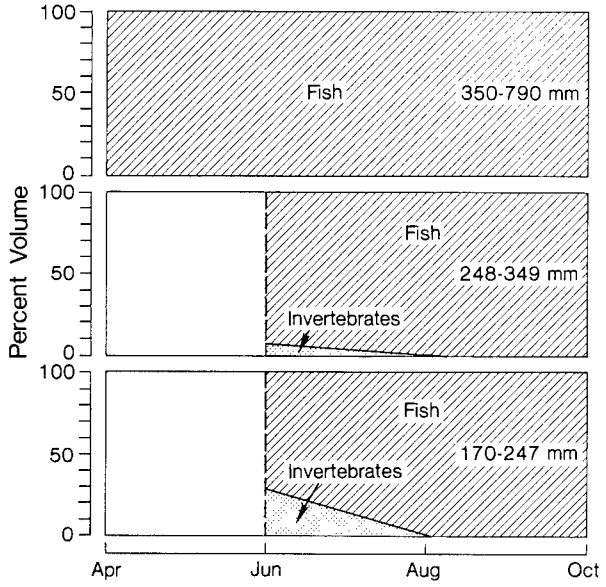

Fig. 21. Changes in major components of walleye diet with season and total length (shown in rectangle at right center of each panel), Tongue River Reservoir, 1980.

in the literature (Burris 1956; Marcy 1954), the levels in the present study were unusually high.

Invertebrates, chiefly zooplankton (Cladocera) and aquatic insects (chironomid larvae and pupae), were also prominent components of the white crappie diet. Invertebrates occurred in about 70-90% of the stomachs of white crappies less than 255 mm long (ages II-IV), and composed 31% of the food volume (Knight 1982); invertebrates continued to occur regularly (frequency, 50-60%) in the stomachs of crappies longer than 255 mm; however, they represented only 4% of the food volume.

Excepting cannibalism, crappies in Tongue River Reservoir generally ate the same kinds of organisms reported for adult crappies from other waters (Marcy 1954; Hoopes 1960; Neal 1961; Keast 1968; Greene and Murphy 1971; Mathur 1972; Baumann et al. 1973), although the relative importance of prey categories differed.

Average and maximum sizes of fish eaten tended to increase with the length of white crappies (Fig. 22), whereas minimum size remained nearly constant because white crappies ate young-of-the-year fish when available. Stomach-content volumes increased markedly as crappie length increased. As in walleyes, much individual variation in stomach-content volume was observed, but large sample sizes compensated for this variation. The frequency of empty stomachs averaged 9% and was unrelated to season or size of fish.

Food of crappies changed significantly between seasons (Fig. 23). Invertebrates dominated in April and June, occurring in 99 and 98% of the stomachs and making up 75 and 85% of the food volumes, respectively (Knight 1982); cladocerans accounted for 25 and 19% and chironomids for 38 and 44% of the food volume in April and June. A radical shift to a predominantly fish diet occurred in

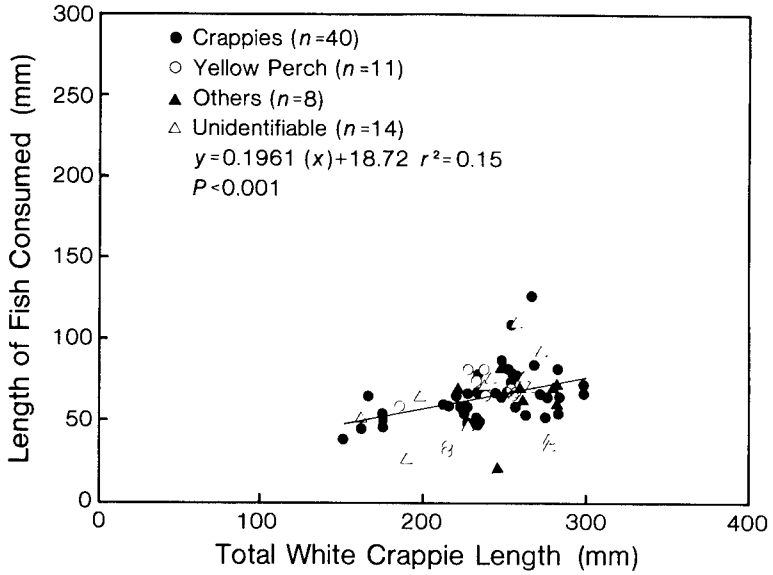

Fig. 22. Estimated total lengths of fish eaten by white crappies as a function of crappie length.

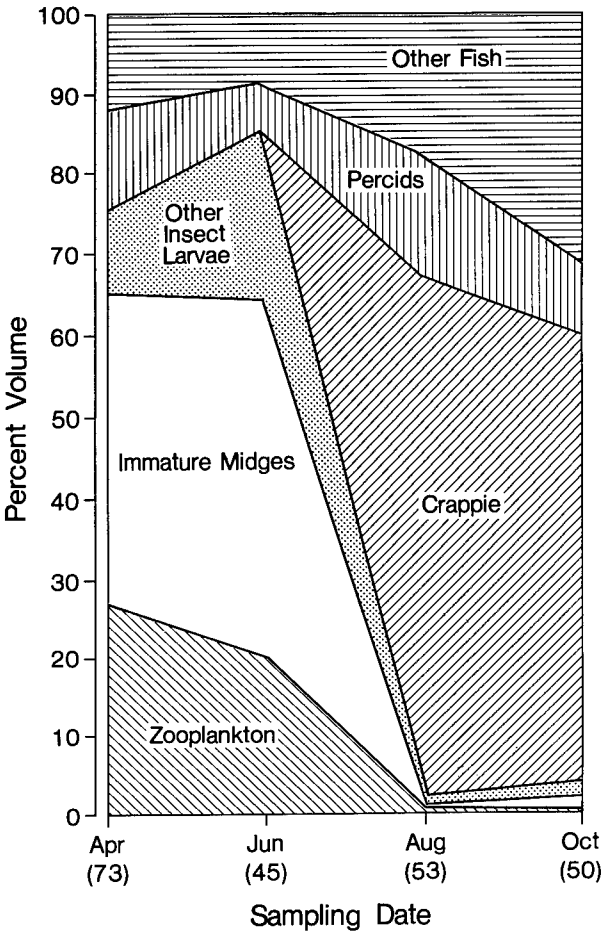

Fig. 23. Seasonal composition of stomach contents of white crappies. Number of white crappies is shown in parentheses beneath each month.

August and continued through October. Fish were in 89 and 76% of the stomachs, and constituted 98 and 96% of the food volume in August and October, respectively. Young-of-the-year crappies were the predominant prey, accounting for 64 and 56% of the food volumes. This seasonal pattern was similar for white crappies of all sizes (Fig. 24), with two exceptions: (1) Large crappies (270 mm long or longer) fed mainly on fish throughout the year, and (2) invertebrates increased slightly in importance from August through October, the increases being greatest in the smallest crappies. This change may have been due to the replacement of young-of-the-year crappies with invertebrates when the young crappies became too large to eat.

Similar seasonal patterns have been reported for crappies in other waters. In Benbrook Lake, Texas, crappies preferred young-of-the-year threadfin shad, but consumed significant amounts of insects when shad were not available (Greene and Murphy 1971). In Conowingo Reservoir, on the lower Susquehanna River, crappies ate mostly fish in fall, but zooplankton and insects were more important in spring (Mathur 1972). These observations again suggested that crappies are opportunistic feeders.

White crappie diets in Tongue River Reservoir also varied diurnally (Fig. 25); percentages of total food volumes contributed by invertebrates were higher during the day, whereas percentages of fish

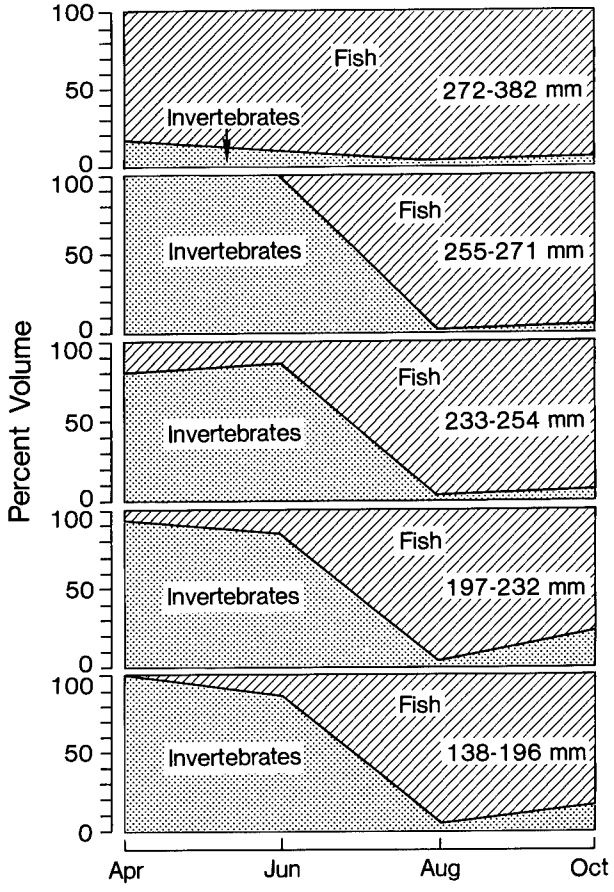

Fig. 24. Seasonal changes in major dietary components of white crappies of differing body length (shown in rectangles at right center of each panel), Tongue River Reservoir, 1980.

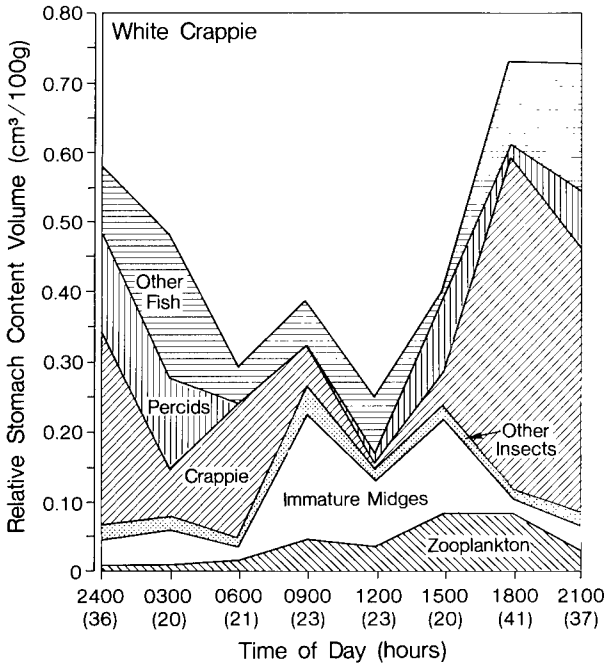

Fig. 25. Diurnal changes in white crappie diet. Number of white crappies shown in parentheses beneath the hour.

were higher at night. Concurrently, the frequency of occurence and the absolute volume of fish eaten during daylight decreased while that of invertebrates increased (Knight 1982).

Daily feeding peaks for crappies in Tongue River Reservoir were generally dawn, near midday, and soon after dark (Fig. 26). Percentages of invertebrates in stomachs increased during midday peaks, whereas dawn and early-evening peaks corresponded with larger percentages of fish. Dawn feeding peaks occurred during seasons when midday peaks were low or nonexistent. In other waters crappies also appeared to eat invertebrates during daytime (Keast 1968; Mathur and Robbins 1972; Baumann et al. 1973) and to eat fish at dawn, dusk, or night (Childers and Shoemaker 1953; Greene and Murphy 1971). White crappies thus appear to feed during hours when their primary forage organisms are most easily captured.

The number and relatively low amplitudes of the observed feeding peaks may result from combining fish of different sizes and diets. Keast (1968), who combined data similarly, observed small, multiple feeding peaks. Greene and Murphy (1971), who also combined fish of different sizes, observed varied feeding patterns. Because the time of feeding is apparently related to both diet composition and body size, feeding patterns should be more distinct among fish of similar sizes.

In summary, young-of-the-year crappies were an important food of both walleyes and white crappies in Tongue River Reservoir. The major differences in the diets of the two species were (1) the

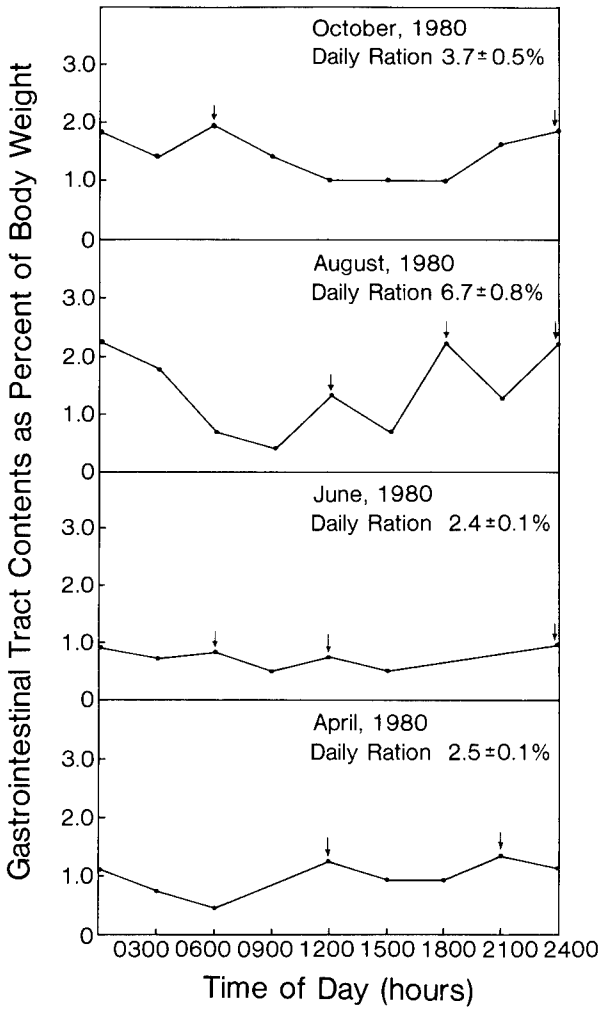

Fig. 26. Patterns of white crappie feeding activity, April-October 1980. Arrows indicate points used to calculate daily ration, expressed as percent of body weight per day.

importance of invertebrates in the diet of white crappies in the spring, and (2) the larger size of forage fishes, other than young-of-the-year crappies, eaten by walleyes. These divergent foraging patterns in the absence of a common dominant prey animal seemed to be related to differences in the sizes of adults of the two species.

Food-consumption Rates

Estimated annual food-consumption rates of walleyes ranged from 0.9 to 3.9% of body weight per day, depending on fish size (Table 17) and combinations of summer and winter activity levels (Table 18). Average daily consumption rate ranged from 1.5 to 2.2% for fish of all size classes (Table 18). Daily maintenance ration for a 1-kg fish was estimated to average 0.7% of body weight (Kitchell et al. 1977) at average monthly water temperatures in Tongue River Reservoir.

Other workers have estimated similar rations for walleyes. Kelso (1972), in laboratory studies on walleyes of ages II-VII, reported daily maintenance rations consistently near 0.5% of body weight. Swenson and Smith (1973) estimated average rations for adult walleyes in Lake of the Woods, Minnesota, at 2.3% (range, 0.5-4.1%) from June to September. Swenson (1977) also compared food of walleyes from several other lakes and estimated that they ate 2.1-2.9% of body weight daily during the growing season, depending on prey densities. Combining Swenson's (1977) and Kelso's (1972) estimates for growing and nongrowing periods, respectively, yielded an annual average of 1.3-1.7%. The model of Kitchell et al. (1977) incorporated these data; thus, it is not surprising that their annual estimate agrees with ours. Both estimates may be low because Swenson and Smith (1973) assumed a linear relation between the amount of food evacuated from the stomach and time; this relation has since been disproved by Elliott and Persson (1978). Given the uncertainties of estimating food-consumption rates, especially indirectly, we estimated the consumption of methylmercury by walleyes with the average range of annual food-consumption rates (1.5-2.2% of body weight per day) for fish of all size-groups.

Estimated food-consumption rate for white crappies in Tongue River Reservoir was 2.5% of body weight per day for April and June; the rate increased to 5.7% in August and decreased to 3.7% by October (Fig. 26). Daily ration estimates exceeded predicted maintenance rations (Kitchell et al. 1977) in all months. Average daily rate of food consumption was estimated to be 2.3±1.2%. We used this range of consumption values during estimations of MeHg consumption. The estimates made in this study are similar to those by Thorpe (1977) and Nakashima and Leggett (1978).

Mathur and Robbins (1972) and Mathur (1972) also reported that feeding activity peaked from June to October; it was moderate in April and May and low from November to March. Greene and Murphy (1971) estimated that minimum food-consumption rates of crappies ranged from 1.6 to 2.8% in late summer. These estimates are probably low because no correction was made for gastric evacuation.

Table 17. *Estimates of annual food-consumption rates (range and median) for different size-classes of walleyes from Tongue River Reservoir. Estimates predicted from body weight, growth rate, and reservoir temperatures (Kitchell et al. 1977). Range is based on possible combinations of activity levels during growing and non-growing periods (summer and winter).*

Walleye size and age		Daily ration (% body weight/day)	
Length (mm)	Estimated age	Range	Median
170-247	I	2.8-3.9	3.35
248-349	II	1.8-2.6	2.20
350-426	III	1.4-2.1	1.75
427-477	IV	1.2-1.8	1.50
478-535	V-VI	1.0-1.6	1.30
>535	≥VII	0.9-1.4	1.15
Average		1.5-2.2	1.85

Mercury in Forage Species

Invertebrates. Total mercury concentrations (μg/g) in invertebrates from Tongue River Reservoir ranged from 0.003 to 0.33 and averaged 0.08 (Table 19). Although sample sizes were small, variability among total mercury concentrations was also relatively small (with the notable exception of the Notonectidae). Our attempts to analyze mercury concentrations in zooplankton were unsuccessful; consequently, we used values from

Table 18. *Estimates of annual food consumption rates for walleyes from Tongue River Reservoir based on possible combinations of growing period and nongrowing period activity levels. Estimates based on the bioenergetics model of Kitchell et al. (1977).*

Walleye size and age		Combination of activity level[a]		Proportion of maximum ration consumed (P)[b]	Daily ration (% body weight/day)
Length (mm)	Estimated age	Winter	Summer		
170-247	I	1	2	0.681	2.8
		1	3	0.823	3.7
		2	2	0.987	2.9
		2	3	1.130	3.9
248-349	II	1	2	0.684	1.8
		1	3	0.840	2.5
		2	2	1.032	1.9
		2	3	1.188	2.1
350-426	III	1	2	0.670	1.4
		1	3	0.832	2.0
		2	2	1.051	1.5
		2	3	1.213	2.1
427-477	IV	1	2	0.657	1.2
		1	3	0.822	1.7
		2	2	1.055	1.3
		2	3	1.220	1.8
478-535	V-VI	1	2	0.648	1.0
		1	3	0.815	1.5
		2	2	1.062	1.1
		2	3	1.229	1.6
>535	VII-XI	1	2	0.646	0.9
		1	3	0.817	1.3
		2	2	1.088	1.3
		2	3	1.259	1.4
Average		1	2	0.664	1.5
		1	3	0.825	2.1
		2	2	1.046	1.6
		2	3	1.207	2.2

[a]Activity levels are numbered as follows: (1) standard metabolic rate as described by Winberg (1956), (2) 2 times the standard metabolic rate as an estimate of the average metabolic rate of adult fish under natural conditions, and (3) 3 times the standard metabolic rate as an estimate of the maximum metabolic rate of adult fish under natural conditions.

[b]Both maximum consumption rates and activity levels are estimates; therefore, P sometimes exceeds 1.0. The calculations in this table are estimates and should not be interpreted as absolute.

the literature to estimate MeHg concentrations in fish diets. Because notonectids were rarely seen in stomachs, we did not use the data for this taxon in subsequent calculations; however, even the mercury concentrations of the two families of Hemiptera differed by an order of magnitude. Differences in their diets may be responsible, as Corixidae are planktivorous and Notonectidae eat other insects.

Table 19. *Mercury concentrations (wet weight) measured in invertebrate foods in Tongue River Reservoir, and for zooplankton reported by other investigators.*

Water, invertebrate taxon, and (for zooplankton) reference[a]	No. of samples	Total mercury (μg/g) Range	Mean	SE
Tongue River Reservoir				
Crustacea	16	0.003–0.20	0.06	0.03
Diptera				
Chironomidae	2	0.066–0.069	0.07	0.02
Hemiptera				
Corixidae	9	0.02–0.04	0.03	0.004
Notonectidae	2	0.21–0.33	0.27	0.78
Various other freshwaters				
Zooplankton				
Sherbin 1979			0.03	
Sherbin 1979			0.010	
Knauer and Martin 1972			0.011	
Trudel et al. 1977			0.019	
Cacoros and Cahn 1973			0.024	
Sherbin 1979			0.025	
Williams and Weiss 1973			0.025	
Armstrong and Hamilton 1973			0.035	
Sherbin 1979			0.040	
Sherbin 1979			0.050	
Flegel 1977			0.060	
Sherbin 1979			0.065	
Sherbin 1979			0.090	
Johnels et al. 1967			0.140	
Sherbin 1979			0.158	
Copeland 1972			0.200	
Mean			0.058	

[a]Sherbin (1979) is a reference for different Canadian freshwaters.

Total mercury concentrations observed in freshwater invertebrates have ranged from 0.002 to 23.2 μg/g (Johnels et al. 1967; Jernelöv and Lann 1971; Armstrong and Hamilton 1973; Cox et al. 1975; Potter et al. 1975; Trudel et al. 1977; Hildebrand et al. 1980). However, invertebrates from uncontaminated waters usually contain less than 0.10 μg/g (Huckabee et al. 1979)—concentrations similar to those for invertebrates from Tongue River Reservoir.

Fishes. Total mercury concentrations were determined in 197 whole forage fish, 40 to 300 mm long, comprising 7 species. The size range closely paralleled the reconstructed total lengths of forage fish found in the stomachs of walleyes and white crappies (21–254 mm). Total mercury concentrations (in μg/g) in forage fish were 0.02–0.40 (crappies, 0.02–0.18; golden shiners, 0.03–0.40; yellow perch, 0.03–0.23; white suckers, 0.03–0.07) and increased with fish length (Fig. 27). Linear regression equations of total length against mercury concentration were similar for all species except golden shiners. The regression equation for the pooled data for all species was used to estimate mercury concentrations in unidentifiable fish remains. All regressions were significantly differ-

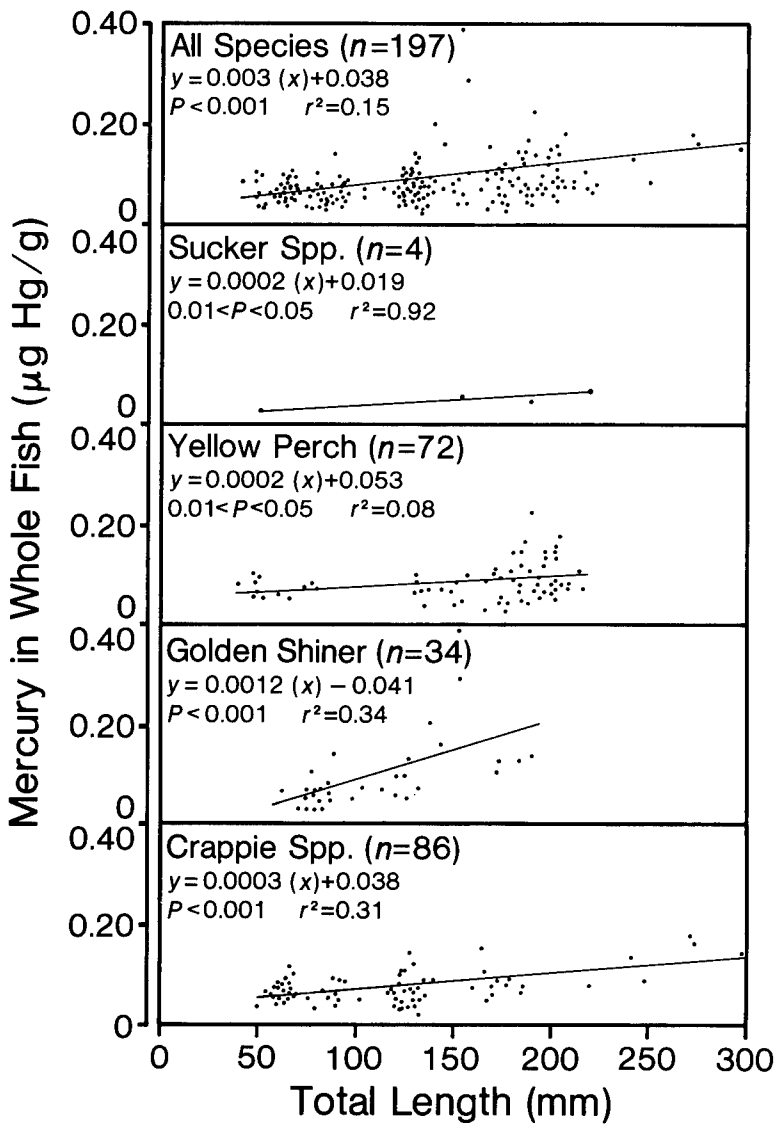

Fig. 27. Relation between concentration (wet weight) of total mercury in whole fish and total length of forage fishes collected from Tongue River Reservoir in 1980.

ent ($P \leq 0.05$); however, coefficients of determination (r^2) were low. Differences in age, sex, species (where combined), and individual behavior probably contributed to this variability (Bache et al. 1971; MacLeod and Pessah 1973; Cross et al. 1973; Prabhu and Hamdy 1977). As in invertebrates, mercury concentrations in forage fish from Tongue River Reservoir generally fell within the ranges observed for these species in uncontaminated waters (Buhler et al. 1973; Gebhards et al. 1973; Potter et al. 1975; Richins and Risser 1975).

Methylmercury in the Diet

Estimated concentrations of MeHg in the diet (µg/g) were 0.023–0.088 (mean, 0.051) for walleyes, and 0.022–0.058 (mean, 0.042) for white crappies, depending on fish size and the assumed percentages of total mercury present as MeHg in dietary components (Table 20). The dietary fractions represented by fish and invertebrates varied with fish size, resulting in different overall MeHg percentages in the diets of fish of different size-classes.

Table 20. *Estimated concentrations (wet weight) of methylmercury in the diets of walleyes and white crappies of different length and ages. Low, mean, and high values correspond to different percentages of MeHg.*[a]

Species and total length (mm)	Age	MeHg (μg/g) in diet		
		Low	Mean	High
Walleye				
170-247	I	0.023	0.039	0.052
248-349	II	0.032	0.049	0.058
350-426	III	0.031	0.047	0.056
427-477	IV	0.032	0.048	0.057
478-535	V-VI	0.032	0.048	0.057
536-790	>VI	0.049	0.075	0.088
Average		0.033	0.051	0.061
White crappie				
138-196	II	0.023	0.038	0.050
197-232	III	0.022	0.038	0.052
233-254	IV	0.022	0.039	0.055
255-271	V	0.032	0.049	0.058
272-382	>V	0.030	0.046	0.055
Average		0.026	0.042	0.054

[a]See text for explanation.

Data on concentrations of mercury, particularly methylmercury, in the diets of fish are scarce. Jernelöv (1972), who measured the mercury concentrations in forage fish eaten by northern pike in a contaminated lake, reported that average mercury concentrations were 5.8 μg/g in the northern pike and 3.1 μg/g in forage fish. Mercury concentrations (μg/g) in whole organisms collected from uncontaminated waters were 1.2 for northern pike, 0.6 for forage fish, and 0.05 for bottom fauna (Jernelöv 1972). Inasmuch as our data suggest that mercury is concentrated in food as it moves through the digestive tract, Jernelöv's (1972) values for forage fish (collected from stomachs) may be high. His value for bottom fauna from an uncontaminated water body, if converted to amount present as MeHg, is similar to our values for MeHg in diets. Norstrom et al. (1976) found MeHg concentrations averaging 0.033 μg/g in the primarily invertebrate diets of yellow perch from Ottawa River. Our values for walleyes and white crappies were slightly higher, probably due to the higher mercury concentrations in the fish component of the diets.

Total mercury concentrations in the stomach and intestinal contents of crappies were generally higher than estimated concentrations in the diet (Table 21); moreover, mercury concentrations were generally higher in intestinal contents than in stomach contents. This difference suggests that mercury was not efficiently assimilated during gastric digestion and was concentrated in the digestive tract. Low mercury assimilation efficiencies (<20%) have been reported for mercury bound in food items under natural conditions (Jernelöv 1968; Phillips and Gregory 1979).

Concentrations of MeHg were consistently 0.01 μg/g higher in the diets of walleyes of ages IV or less (<477 mm long) than in the diets of white crappies of the same estimated ages. The MeHg content of the diets of both species increased sharply at certain ages, probably in response to observed increases in the amount and size of the fish eaten (Knight 1982). One such shift occurred between ages IV and V in crappies, and a larger shift between ages VI and VII in walleyes. Increases in dietary MeHg concentrations also coincided with increases in the rates of mercury accumulation with age (Fig. 18).

Other workers have also observed higher mercury concentrations or higher percentages of MeHg in fish at higher trophic levels (Armstrong and Hamilton 1973; Huckabee et al. 1974; Potter et al. 1975; Richins and Risser 1975; Kendall 1978; Cox et al. 1979; Meister et al. 1979). These findings have often been related to differences in feeding habits and considered as evidence of food chain biomagnification. Two valid criticisms of this reasoning were offered by deFreitas et al. (1977) and Huckabee et al. (1979): (1) Because predators live longer than most prey species, their exposure to mercury is greater, and they attain higher mercury concentrations regardless of trophic magnification; and (2) most investigators have not accounted for mercury dilution by growth, which results in lower mercury concentrations in faster growing species. These authors stated that organisms occupying lower trophic levels usually grow faster than those at higher trophic levels, causing trophic effects to be exaggerated. However, neither argument applies in the present study. First, we compared fishes of similar ages; second, the daily growth rates of walleyes exceeded those of white crappies and thus would tend to mask rather than exaggerate trophic effects. Thus food-chain transport probably accounts for the differences in rates of mercury uptake between walleyes and white crappies in Tongue River Reservoir.

Table 21. *Mercury concentrations (wet weight) in stomach and intestinal contents of white crappies from Tongue River Reservoir.*

Total length (mm)		Date	Stomachs				Intestines			
				Hg(μg/g)		Estimated MeHg[a]		Hg (μg/g)		Estimated MeHg[a]
Interval	Mean	(1980)	*n*	Mean	SE	(μg/g)	*n*	Mean	SE	(μg/g)
138-196	166	August	3	0.086	0.037	0.063	2	0.053	0.045	0.039
		October	4	0.043	0.035	0.031	1	0.161	—	0.118
		Combined	7	0.062	0.029	0.045	3	0.079	0.137	0.058
197-232	224	May	5	0.051	0.024	0.035	6	0.077	0.035	0.053
		August	9	0.090	0.013	0.062	7	0.093	0.047	0.059
		October	3	0.071	0.057	0.049	2	0.145	0.925	0.099
		Combined	17	0.075	0.013	0.051	15	0.094	0.029	0.064
233-254	244	May	9	0.040	0.019	0.026	8	0.065	0.014	0.043
		August	7	0.089	0.050	0.059	6	0.067	0.035	0.044
		October	11	0.098	0.047	0.064	8	0.073	0.030	0.048
		Combined	27	0.076	0.023	0.050	22	0.068	0.012	0.045
255-271	263	May	1	0.074	-	0.061	1	0.074	-	0.061
		August	2	0.099	0.296	0.082	2	0.059	0.099	0.049
		October	11	0.081	0.021	0.067	9	0.106	0.061	0.088
		Combined	14	0.083	0.018	0.071	12	0.095	0.045	0.079
>271	283	October	0	-	-	-	1	0.033	-	0.027

[a]Methylmercury concentration was estimated by dividing the total mercury concentration into fractions attributable to invertebrates and fish (Table 14), multiplying each by the appropriate mean percent MeHg (from literature), and summing the resulting values.

Methylmercury Accumulation by Fish

We estimated MeHg accumulation from food by walleyes and white crappies from food-consumption rates and dietary concentrations of MeHg. These estimates were compared with observed MeHg accumulations, and a theoretical framework was developed for assessing the relative importance of food as a source of MeHg to fish.

Observed accumulations. Total mercury concentrations in walleyes and white crappies were less in 1980 than in fish of the same ages in 1978; however, the amount and concentration of mercury in individual cohorts increased from 1978 to 1980 (Table 22). For example, in 1978, walleyes of age II averaged 0.13 μg/g (representing 34 μg/g of Hg), whereas in 1980 the same fish (at age IV) averaged 0.175 μg/g (representing 155 μg of Hg). However, walleyes of age II in 1980 averaged only 0.08 μg/g (22 μg of Hg). The quantity of mercury in the 1976 cohort increased by 121 μg (60 μg/year), despite the corresponding decline in mercury concentration in

Table 22. *Observed annual rate of methylmercury accumulation* dM/dt *from 1978 to 1980. Low, mean, and high values of* dM/dt *correspond to different percentages of MeHg.*

Species and length (mm)	Age	Observed annual MeHg accumulation rate *dM/dt* (μg)		
		Low	Mean	High
Walleye				
170-247	I	2.8	4.2	4.9
248-349	II	6.2	9.4	11
350-426	III	13	19	23
427-477	IV	34	51	60
478-535	V-VI	43	65	76
536-790	>VI	194	294	346
White Crappie				
138-196	II	1.5	2.2	2.6
197-232	III	4.9	7.4	8.8
233-254	IV	5.5	8.3	9.8
255-271	V	2.6	4.0	4.7
272-382	VI-VIII	5.1	7.8	9.2

fish of the same age. Correction for the percentage MeHg yielded the values shown in Table 22. Increases between years were smaller than suggested by the values from either year alone (i.e., the difference between fish of age IV and II from the same year).

Mercury concentrations in Tongue River Reservoir fishes were elevated in 1979, relative to 1978 (Phillips 1979) and 1980, indicating that the amount of methylmercury available to fish may change from one year to the next, and that differences between years can result from fluctuating environmental conditions. Jernelöv et al. (1975) indicated that mercury in top predators does not reach equilibrium with that in the environment for 10 to 15 years after changes in their MeHg exposure regime. Thus mercury fluxes in highly dynamic systems, such as reservoirs, may never attain equilibrium conditions.

The MeHg accumulated by walleyes in 1 year increased rapidly with age, whereas the yearly accumulation of MeHg by white crappies increased through age IV but was slower in fish of ages V–VIII. Rates of accumulation in walleyes also slowed at about the same age (V–VI), but the reason for this decrease is unknown. Walleyes accumulated more MeHg at any given age than did white crappies. Accumulation rates seemed to be positively related to both age and trophic level.

Uptake from food. Low assimilation coefficients in the range of reported values gave the only values of dF/dt (annual uptake from food) that were less than observed accumulation rates (dM/dt). This suggests that 0.15 is a realistic assimilation coefficient for Tongue River Reservoir fish. Phillips and Gregory (1979) showed that dietary MeHg assimilation was low in natural situations due to the exposure regimes of food items

Table 23. *Calculated annual dietary uptake of methylmercury by walleyes in Tongue River Reservoir based on a range of food-consumption rates and methylmercury concentrations in the diet. An assimilation efficiency for dietary methylmercury of 0.15 was used for all calculations.*

Length (mm)	Age	MeHg in diet (μg/g food)		Annual ration (R) (g food/g fish)		MeHg uptake from food (dF/dt; μg/year)
170–247	I	Low	(0.02)	Low	(5.5)	0.0
		Mean	(0.04)	Median	(6.8)	1.8
		High	(0.05)	High	(8.0)	2.9
248–349	II	Low	(0.03)	Low	(5.5)	4.7
		Mean	(0.05)	Median	(6.8)	8.9
		High	(0.06)	High	(8.0)	12.4
350–426	III	Low	(0.03)	Low	(5.5)	9.4
		Mean	(0.05)	Median	(6.8)	17.6
		High	(0.06)	High	(8.0)	24.7
427–477	IV	Low	(0.03)	Low	(5.5)	17.7
		Mean	(0.05)	Median	(6.8)	33.0
		High	(0.06)	High	(8.0)	46.2
478–535	V–VI	Low	(0.03)	Low	(5.5)	28.7
		Mean	(0.05)	Median	(6.8)	53.8
		High	(0.06)	High	(8.0)	75.2
536–790	>VI	Low	(0.05)	Low	(5.5)	72.1
		Mean	(0.08)	Median	(6.8)	135
		High	(0.09)	High	(8.0)	189

(low concentrations over long periods), which result in the binding of MeHg to relatively non-digestible food constituents. High assimilation values are frequently observed in the laboratory (Suzuki and Hatanaka 1975; Sharpe et al. 1977), where conditions generally favor loose binding of MeHg to food. This conclusion is supported by studies that indicated that MeHg is assimilated into the blood and later redistributed to other, less digestible tissues, especially muscle components (Giblin and Massaro 1973; Olson et al. 1973; Laarman et al. 1976; McKim et al. 1976). Consequently, only low assimilation efficiencies are presented or used in the following calculations.

The amount of MeHg assimilated annually from food increased with size (and age) in both walleyes and white crappies (Tables 23 and 24). Uptake values were consistently higher in walleyes than in crappies of the same age. These trends resulted from differences in the MeHg content of the diet of these fishes. Many investigators have shown that mercury accumulation rates increase with increasing concentrations in food (Miettinen 1975; Lock 1975; Wobeser 1975; Huckabee et al. 1978). It has been hypothesized that increases in the amount of MeHg consumed (and assimilated) with increasing size and trophic level account for differences in MeHg accumulation under natural conditions; however, such increases have not been previously documented (Richins and Risser 1975; Benson et al. 1976; Phillips et al. 1980). Our results indicated that MeHg uptake from food does indeed increase with size and trophic level.

Fraction attributed to food. The percentage of accumulated MeHg attributable to food ranged from about 10% to more than 100% in both walleyes and white crappies, depending on the combination of *dF/dt*, *dM/dt*, and *dE/dt* used in the estimation. Obviously, only one combination yielding a total less than 100% is possible. Age and species trends from the two methods of calculating *FF* (the fraction derived from food) were similar; however, for comparisons, the more rigorous method—equation (2)—gave the higher values.

Table 24. *Calculated annual dietary uptake of methylmercury by white crappies in Tongue River Reservoir based on a range of food-consumption rates and methylmercury concentrations in the diet. An assimilation efficiency for dietary methylmercury of 0.15 was used for all calculations.*

Length (mm)	Age	MeHg in diet (μg/g food)		Annual ration (*R*) (g food/g fish)		MeHg uptake from food (*dF/dt*; μg/year)
138-196	II	Low	(0.02)	Low	(4.0)	0.4
		Mean	(0.04)	Mean	(8.4)	1.4
		High	(0.05)	High	(12.8)	2.9
197-232	III	Low	(0.02)	Low	(4.0)	1.4
		Mean	(0.04)	Mean	(8.4)	5.0
		High	(0.05)	High	(12.8)	10.5
233-254	IV	Low	(0.02)	Low	(4.0)	2.3
		Mean	(0.04)	Mean	(8.4)	8.4
		High	(0.06)	High	(12.8)	17.7
255-271	V	Low	(0.03)	Low	(4.0)	4.1
		Mean	(0.05)	Mean	(8.4)	13.0
		High	(0.06)	High	(12.8)	23.7
272-382	VI-VIII	Low	(0.03)	Low	(4.0)	5.9
		Mean	(0.05)	Mean	(8.4)	19.0
		High	(0.06)	High	(12.8)	34.5

This difference was more pronounced in white crappies than in walleyes because the *dM/dt* values for crappies were much smaller than those predicted by the 1980 data.

When mean values for all variables except assimilation were used (*dF/dt*—low-mean; *dM/dt*—mean; *dE/dt*—mean), the percentages of accumulated MeHg attributable to food were 27–83 in walleyes and 21–91 in white crappies, depending on size and method of estimation. For the two largest size-groups of crappies, only low values of *R* (ration) and *C* (MeHg in diet) produced *FF* less than 1 with equation (2); consequently, low-low values of *dF/dt* were used for these two size-groups. In white crappies, the fraction of MeHg derived from food generally increased with size and age. These data support the hypothesis that the higher mercury concentrations in older fish resulted from increased exposure through food as well as from longer exposure times. The walleye data were less conclusive; the fraction derived from food was lowest among both the smallest and the largest size-groups. In older fish, accumulation rates increased as fast or faster than rates of uptake from food, and the fraction derived from food remained stable or declined slightly. Although uptake from water may have increased with size, an alternate explanation for these observations might be provided by further study of seasonal and yearly changes in the uptake of MeHg from food.

Average percentages of accumulated MeHg from food (based on mean variables) were 41 and 62 for walleyes and 51 and 73 for white crappies for equations (1) and (2), respectively. Unfortunately, the uncertainty associated with these estimates is large, and any extrapolation of the data—especially specific percentages—must be approached cautiously. Nonetheless, the data showed that under realistic conditions food was a major source of accumulated MeHg in Tongue River Reservoir fishes. Our results emphasize the difficulties involved in estimating pathways of mercury uptake and illustrate the need for better quantification of many of the variables not measured. With some exceptions (Hannerz 1968; Fagerström and Åséll1976), most other investigators have also concluded that, under certain (variable) conditions, food can be a significant source of MeHg to aquatic organisms (Colwell et al. 1975; Huckabee et al. 1975; Jernelöv et al. 1975; Lock 1975; Huckabee et al. 1978; Hildebrand et al. 1980; Ribeyre et al. 1981). The question then becomes, How significant? This and other studies have failed to accurately determine the importance of food, primarily due to the analytical difficulties of measuring MeHg in food organisms. Like MeHg concentrations in water, MeHg concentrations in food organisms in natural systems often are below current detection limits. Since many factors affect the rate and efficiency of MeHg uptake by fishes from both food and water, their relative contributions probably vary.

Relation of Limnology of Three Reservoirs to Mercury Accumulation by Fish

Trends in limnological characteristics, including bacterial densities in Tongue River, Cookson, and Nelson reservoirs are depicted by surface and bottom graphs in Figs. 28–33, and densities by isopleths (bacteria excepted) for the deepest station in each reservoir in Figs. 34–36. In general, the reservoirs are shallow, well-mixed systems with similar temperature regimes, although Tongue River Reservoir is somewhat deeper and stratifies to some extent. The surface waters have similar dissolved oxygen concentrations and redox potentials. Variability between months and stations—particularly in conductivity and pH—was greater in Tongue River Reservoir than in Cookson and Nelson reservoirs. The isopleths show that Cookson and Nelson reservoirs were well mixed, even at the deepest stations. Statistical comparisons by multifactor analysis of variance ($P \leq 0.01$; Snedecor and Cochran 1967) of each variable by reservoir, month, and depth are shown in Table 25. Redox potentials at the bottom differed significantly among the reservoirs at their deeper stations ($P = 0.01$).

The concentration of total mercury in water from all three reservoirs was consistently low, averaging 0.01 μg/L (SD, 0.005) and ranging from 0.01 to 0.03 μg/L. Total mercury concentrations (mean, 0.016 μg/L) in water of Tongue River Reservoir and its inflow and outflow were significantly higher ($P = 0.05$) than in the two other reservoirs—Nelson (0.012 μg/L) and Cookson (0.011 μg/L).

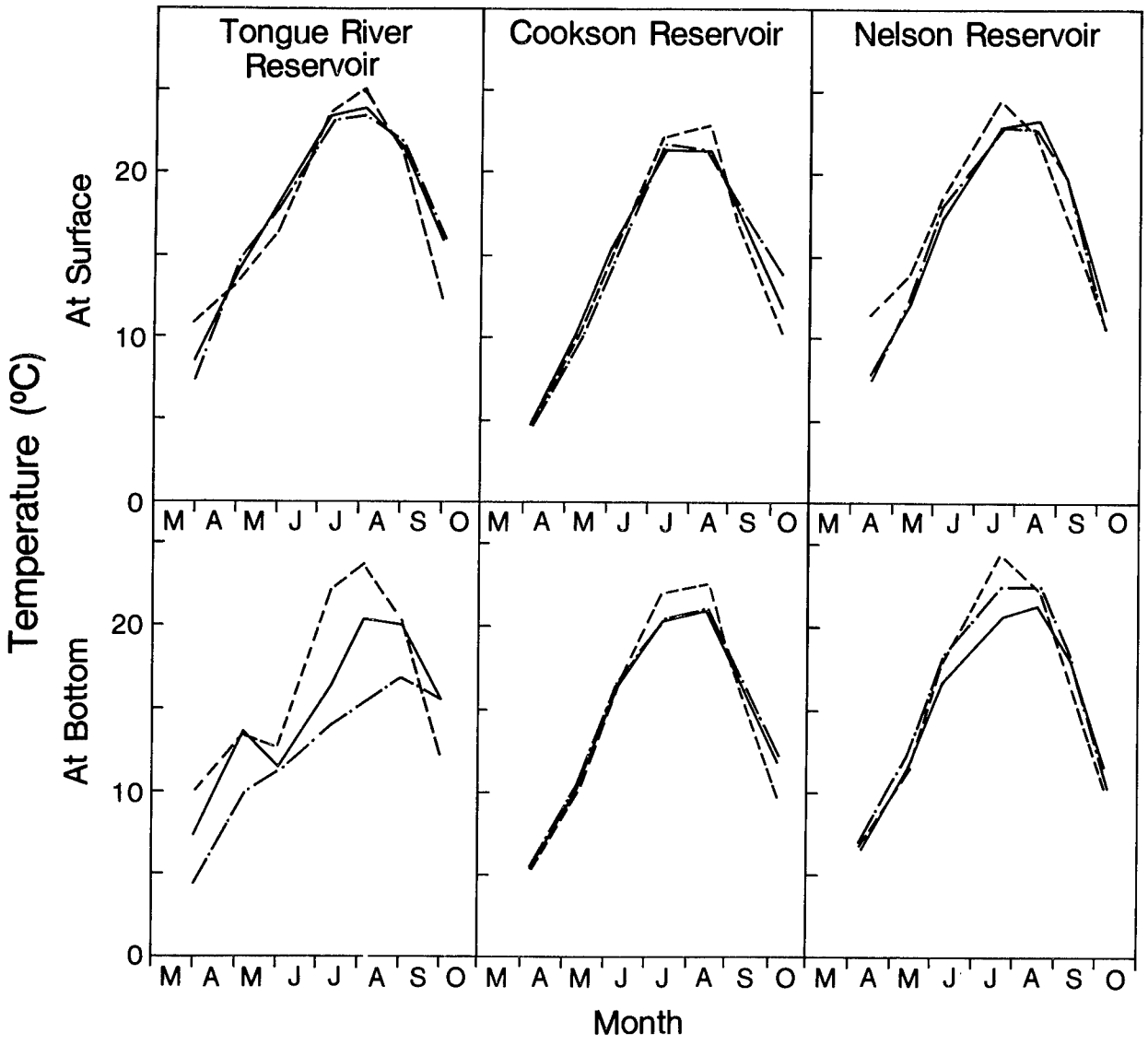

Fig. 28. Seasonal trends in temperature of surface and bottom water for three reservoirs at their upper (- - -), middle (———), and lower (— · — · —) stations.

Bacterial densities in the inflows and outflows of the reservoirs (seven samples per stream) indicated a fourfold greater density in the inflow to Cookson Reservoir (125 ± 75 × 10^4 bacteria/mL; mean ± SD) than in the other streams (Cookson outflow 16 ± 9; Nelson inflow 33 ± 18, outflow 17 ± 10; Tongue River inflow 35 ± 11, outflow 28 ± 15).

Sediment analyses indicated that the substrate of the reservoirs differed considerably (Table 26). Total phosphorus and sulfur concentrations were significantly higher ($P = 0.01$) in Tongue River Reservoir than in the other two reservoirs, and the odor of hydrogen sulfide was detectable in its sediments.

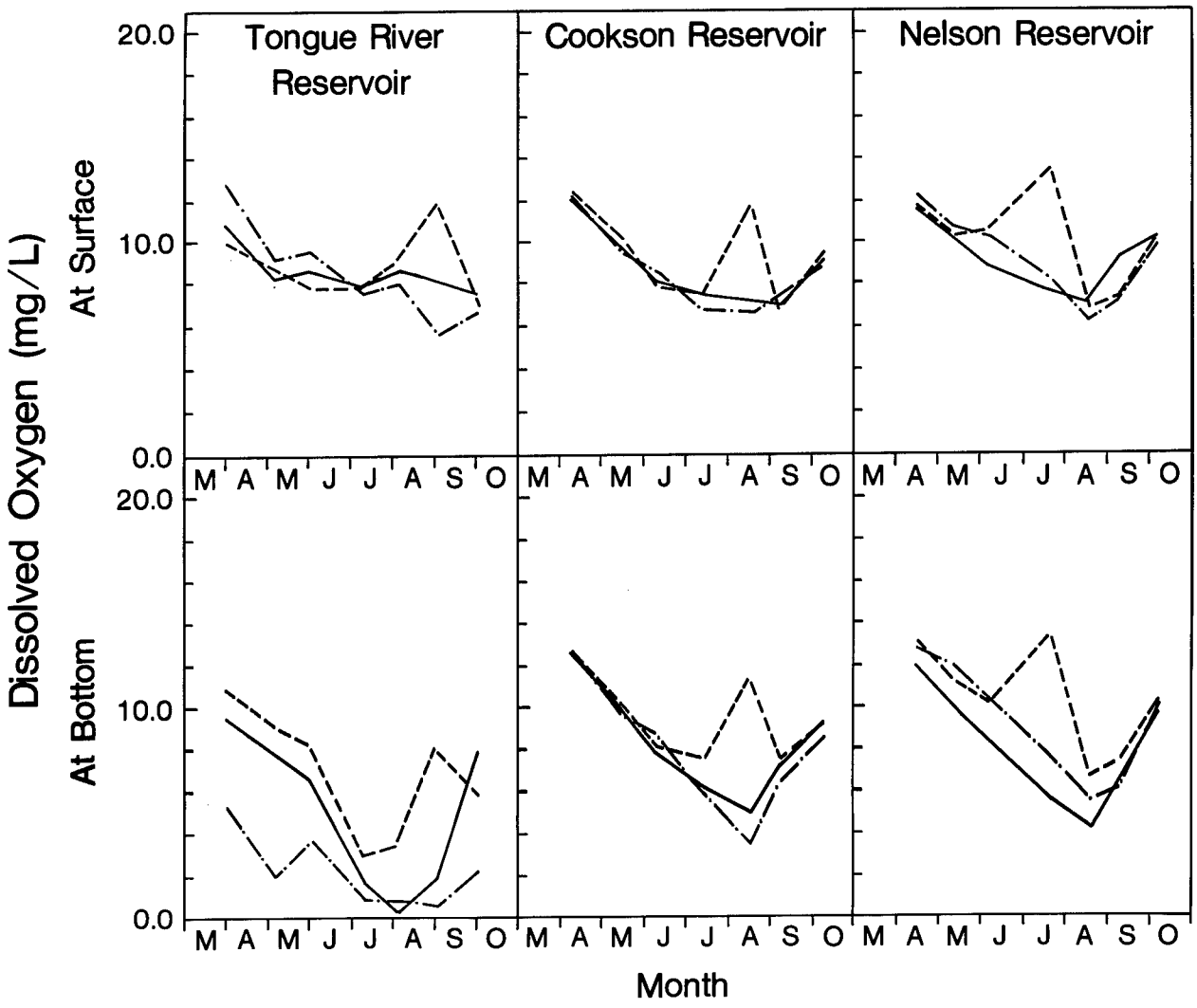

Fig. 29. Seasonal trends in dissolved oxygen of surface and bottom water for three reservoirs at their upper (- - -), middle (———), and lower (— · — · —) stations.

Sediments

Dissolved oxygen gradients and consequently redox potential gradients that occur in sediments result in sediment layers with different capacities for mercury methylation (Bartlett and Craig 1981). Under extremely reducing conditions (< -100 mV), sulphide chemistry predominates; that is, although mercury is being methylated, complexation by sulfur reduces its biological availability. Although demethylating processes reduce methylmercury concentrations under strongly oxidizing conditions (>150 mV), mildly oxidizing conditions ($-100 \leq$ mV ≤ 150) seem to be most conducive to making methylmercury available to biota. Since E_h declines rapidly with sediment depth, reservoirs with aerobic bottom waters have a sediment zone in the E_h range most conducive to methylmercury bioavailability. During part of the year in anaerobic regions of Tongue River Reservoir (and to some extent, Nelson Reservoir) the E_h of the surface sediment is probably below -100 mV. Cookson Reservoir, because its bottom waters are aerobic throughout summer and fall, appears to have the most favorable conditions for bioavailability of methylmercury; the faster rate of mercury uptake by walleyes in this reservoir supports this hypothesis.

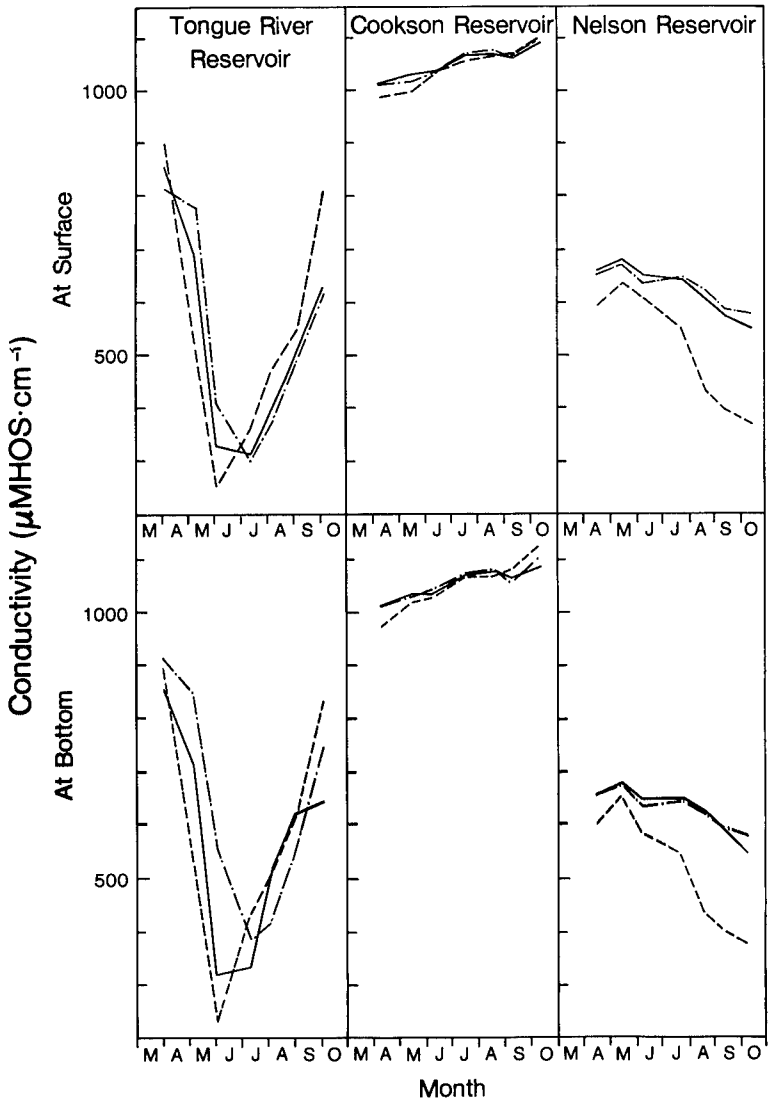

Fig. 30. Seasonal trends in specific conductance of surface and bottom water for three reservoirs at their upper (· · ·), middle (———), and lower (— · — · —) stations.

Factors that limit mercury availability and that influence bacterial activity largely determine the rate of mercury methylation at the sediment-water interface. Availability of nutrients—including nitrogen, phosphorus, and organic matter—tend to stimulate methylmercury production (Jackson and Woychuk 1980). However, the combined presence of organic carbon (indicative of humic matter) and colloidal iron oxides—the situation in Tongue River Reservoir—tends to inhibit methylmercury availabilty (Rust 1977). Sediments with lower ratios of iron oxide to manganese oxide, such as those observed in Cookson Reservoir, can increase the bioavailability of methylmercury (Jackson and Woychuk 1980). This observation is consistent with the high mercury concentrations observed in Cookson Reservoir walleyes.

The physical composition of sediments influences the amount of surface area available for

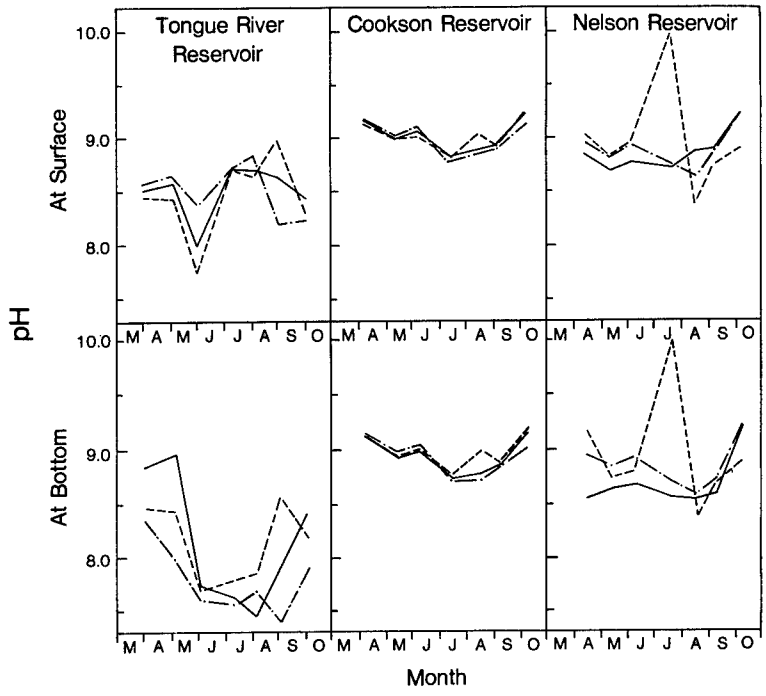

Fig. 31. Seasonal trends in pH of surface and bottom water for three reservoirs at their upper (- - -), middle (———), and lower (— · — · —) stations.

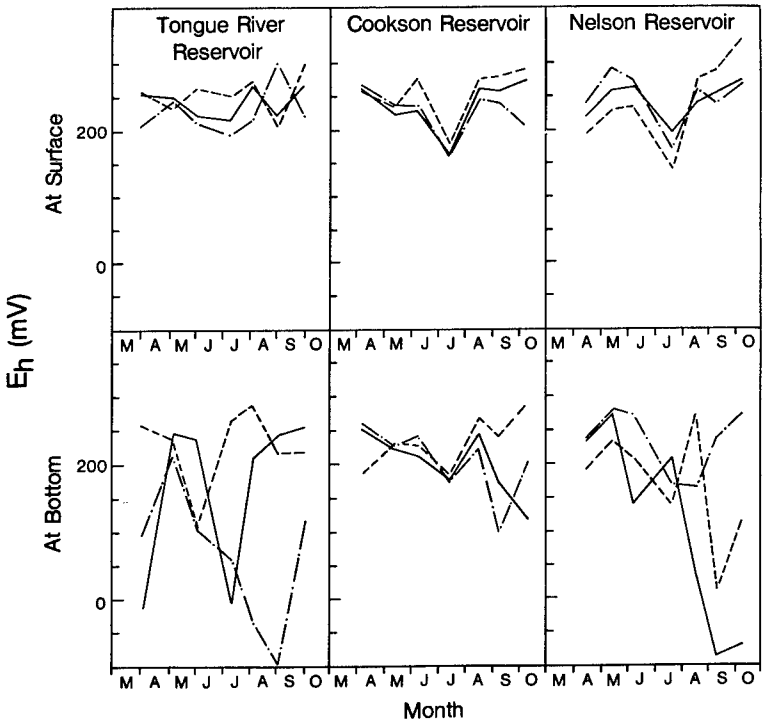

Fig. 32. Seasonal trends in oxidative reductive potential (E_h) of surface and bottom water for three reservoirs at their upper (- - -), middle (———), and lower (— · — · —) stations.

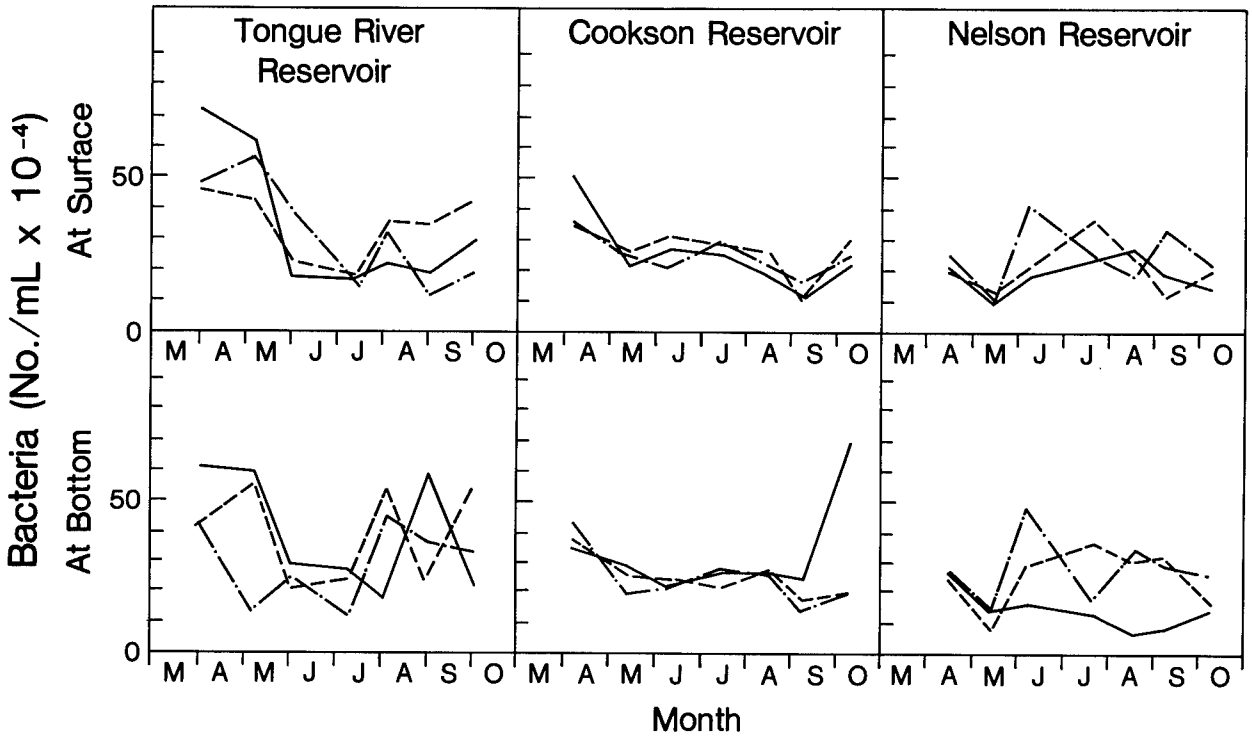

Fig. 33. Seasonal trends in density of bacteria in surface and bottom water for three reservoirs at their upper (- - -), middle (———), and lower (— · — · —) stations.

mercury binding and also affects permeability to oxygen diffusion. Total mercury concentrations in our sediments paralleled the percentages of clay present. Bartlett and Craig (1981) also found positive correlations of total mercury and methylmercury concentrations with percent silt (and clay, diameter ≤ 0.0625 mm). Furutani and Rudd (1980) noted higher rates of mercury methylation in sediment from a lake having a lower total mercury concentration and a lower percentage of clay than was present in Clay Lake, Ontario. A similar comparison can be made between Tongue River Reservoir (low methylmercury bioavailability) and Cookson Reservoir (high methylmercury bioavailability).

Macrobenthos

Our study indicated distinct differences in macroinvertebrate densities among reservoirs. Densities of macroinvertebrates are affected by several factors (Cowell and Hudson 1967). Water temperature influences the rate of development and population turnover, and low dissolved-oxygen concentrations and sandy substrates limit populations. Water-level fluctuations and wind and wave action on the bottom also inhibit colonization and can result in emigration. In our reservoirs, low dissolved oxygen in Tongue River Reservoir and the sand substrate in Cookson may have limited benthos densities.

Benthic invertebrates can influence mercury uptake by fishes in several ways (Petr 1977; Boddington et al. 1979). Bioturbation (physical disturbance of sediments by biota) releases methylmercury to the water column and suspends particles that act as sites for methylation. Burrowing invertebrates circulate water within the sediments, facilitating the release of methylmercury and resulting in a larger zone of methylmercury production. Invertebrates also absorb both inorganic and organic mercury (Bissonette 1977; Miller 1977; Trudel 1977; Guthrie and Cherry 1979) and, when preyed upon by fish, provide a food source of mercury (Qadri and Rodgers 1977). Although our benthic sampling was extremely limited, the data indicated that Nelson Reservoir had the highest potential for organism-mediated water

Table 25. *Means and significant differences (multifactor* ANOVA; $P \leq 0.01$; *seven samples per mean) of bacterial densities and chemical and physical characteristics of Tongue River Reservoir, Cookson Reservoir, and Nelson Reservoir, by reservoir, month, and depth.*[a]

Category: reservoir, month, or depth stratum	Characteristic					
	Dissolved oxygen (mg/L)	Temperature (°C)	pH	E_h (mV)	Conductivity (μmhos/cm)	Bacterial density (No. × 10^4/mL)
Reservoir						
Tongue River	6.9*	16.6	8.22*	213	550*	33.8*
Cookson	8.8	14.8*	8.99*	230	1,055*	26.8*
Nelson	9.3	15.9	8.85*	214	600*	20.4*
Month						
April	11.8*	6.8*	$8.85^{y,z}$	$222^{y,z}$	851^{z}	40.3*
May	9.4^{w}	12.0^{w}	$8.74^{y,z}$	242^{x}	$813^{y,z}$	29.1^{w}
June	8.4^{w}	15.8*	8.54^{w}	228^{w}	668^{w}	26.1^{w}
July	6.5^{x}	21.4^{x}	$8.57^{w,x}$	$181^{w,x}$	$691^{w,x}$	22.5^{w}
August	6.5^{x}	22.1^{x}	$8.55^{w,x}$	$232^{w,x}$	$706^{w,x}$	27.4^{w}
September	6.9^{x}	18.9*	$8.67^{w,x,y}$	$202^{w,x}$	732^{x}	22.1^{w}
October	8.6^{w}	12.6^{w}	8.86^{w}	$228^{w,x}$	793^{y}	25.4^{w}
Depth						
Surface	9.0	16.4	8.81	242	728	26.9
Mid-depth	8.4	16.0	8.67	243	751	26.7
Bottom	7.4*	14.8*	8.56*	179*	778	28.7

[a]Asterisk indicates that differences from other values in the same category and column are highly significant ($P \leq 0.01$); among values for different months, entries within a column bearing the same superscripts are not significantly different.

exchange and release of methylmercury to the water. Cookson Resrvoir ranked next, followed by Tongue River Reservoir.

Water Column

Methylation of mercury in the water column is affected by many factors, including microbial activity, nutrient supply (thus, the level of primary production), availability of mercury, and degree of thermal stratification (Furutani and Rudd 1980; Topping and Davies 1981).

Water temperatures did not differ appreciably among the three reservoirs of this study. However, temperature may be a factor in the generally higher mercury content in fish in reservoirs than in rivers. The well-mixed conditions of the present reservoirs ensure that relatively homogeneous temperatures occur throughout the water column. High water temperatures favor methylation activity at the sediment-water interface.

All three reservoirs were aerobic throughout most of the water column during the ice-free portion of the year, although bottom waters in Tongue River Reservoir were nearly anaerobic (Fig. 34). Wind-generated mixing maintained nearly isograde dissolved oxygen profiles in Nelson and Cookson reservoirs, and river currents limited anaerobic conditions in Tongue River Reservoir. Supersaturated oxygen conditions, indicative of phytoplankton blooms and high nutrient concentrations, occurred in late summer in all three reservoirs. These blooms may have stimulated methylation of mercury by providing organic substrates for microbial growth (Furutani and Rudd 1980) and could be a factor in elevated mercury in fishes. The nearly anaerobic conditions in Tongue River Reservoir apparently caused sulfur reduction, as evidenced by the distinct hydrogen sulfide odor in the sediments. Mercury binds to sulfur, and mercury bound as HgS is not readily methylated in anoxic environments. This rela-

Table 26. *Means of chemical and physical components of sediments at stations in Tongue River, Nelson, and Cookson reservoirs. All values are percentages, except total mercury, which is expressed in μg/g.*

Reservoir and station	Total nitrogen	Total phosphorus	Total sulfur	Total iron	Total manganese	Extractable iron	Extractable manganese	Total mercury	Ash	Sand[a]	Silt[b]	Clay[c]
Tongue River												
Upper	0.05	0.10	0.12	2.43	0.05	0.15	0.03	0.070	94.3	29.7	51.9	18.4
Middle	0.24	0.10	0.31	3.50	0.07	0.31	0.05	0.099	90.5	6.6	30.2	63.2
Lower	0.26	0.12	0.35	3.63	0.08	0.53	0.06	0.145	90.1	2.6	24.7	72.7
Mean	0.18	0.11	0.26	3.19	0.07	0.33	0.05	0.105	91.6	13.0	35.6	51.4
Nelson												
Upper	0.10	0.07	0.11	2.69	0.03	0.24	0.02	0.077	94.6	16.9	42.9	40.2
Middle	0.20	0.06	0.07	3.61	0.05	0.17	0.03	0.144	91.8	4.5	37.0	58.5
Lower	0.22	0.04	0.12	1.77	0.01	0.16	0.003	0.082	94.7	30.7	50.9	18.4
Mean	0.17	0.06	0.10	2.69	0.03	0.19	0.02	0.101	93.7	17.4	43.6	39.0
Cookson												
Upper	0.39	0.04	0.18	2.86	0.06	0.10	0.04	0.109	88.9	21.2	48.9	29.9
Middle	0.22	0.08	0.10	1.64	0.05	0.14	0.03	0.06	93.4	72.9	19.1	8.0
Lower	0.06	0.05	0.06	2.20	0.06	0.07	0.03	0.050	96.1	46.0	36.4	17.6
Mean	0.22	0.06	0.11	2.23	0.06	0.09	0.03	0.073	92.8	46.7	34.8	18.5

[a]Diameter > 0.0625 < 2mm.
[b]Diameter > 0.0039 < 0.0625 mm.
[c]Diameter > 0.004 mm.

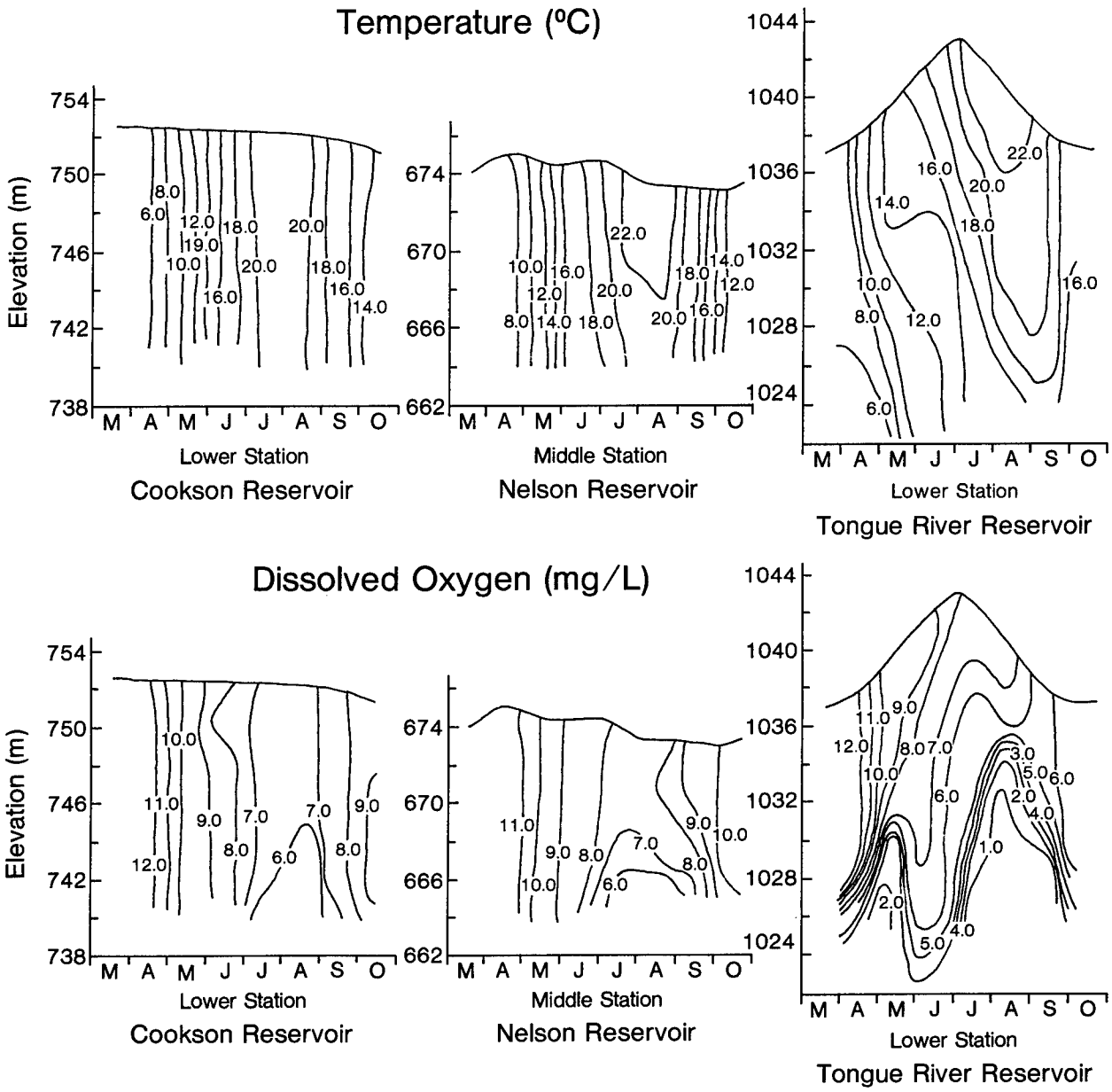

Fig. 34. Seasonal temperature (*upper*) and dissolved oxygen (*lower*) isopleths for the deepest station at Nelson, Cookson, and Tongue River reservoirs.

tion could help to explain the lower mercury concentrations in fish from Tongue River Reservoir.

In surface waters of the three reservoirs, redox potentials did not differ significantly and were not a factor in determining differences in mercury concentrations in fish. However, in Tongue River Reservoir, and occasionally in Nelson Reservoir, the low (< zero) redox potential of bottom waters probably resulted in complexation of mercury by sulphide. This observation is consistent with observations of mercury in fish from the three locations.

The pH of all three reservoirs was relatively high, ranging from 7.2 to 10.0 (mean, 8.6); and thus indicative of well-buffered systems. High pH values coincided with periods of high rates of pho-

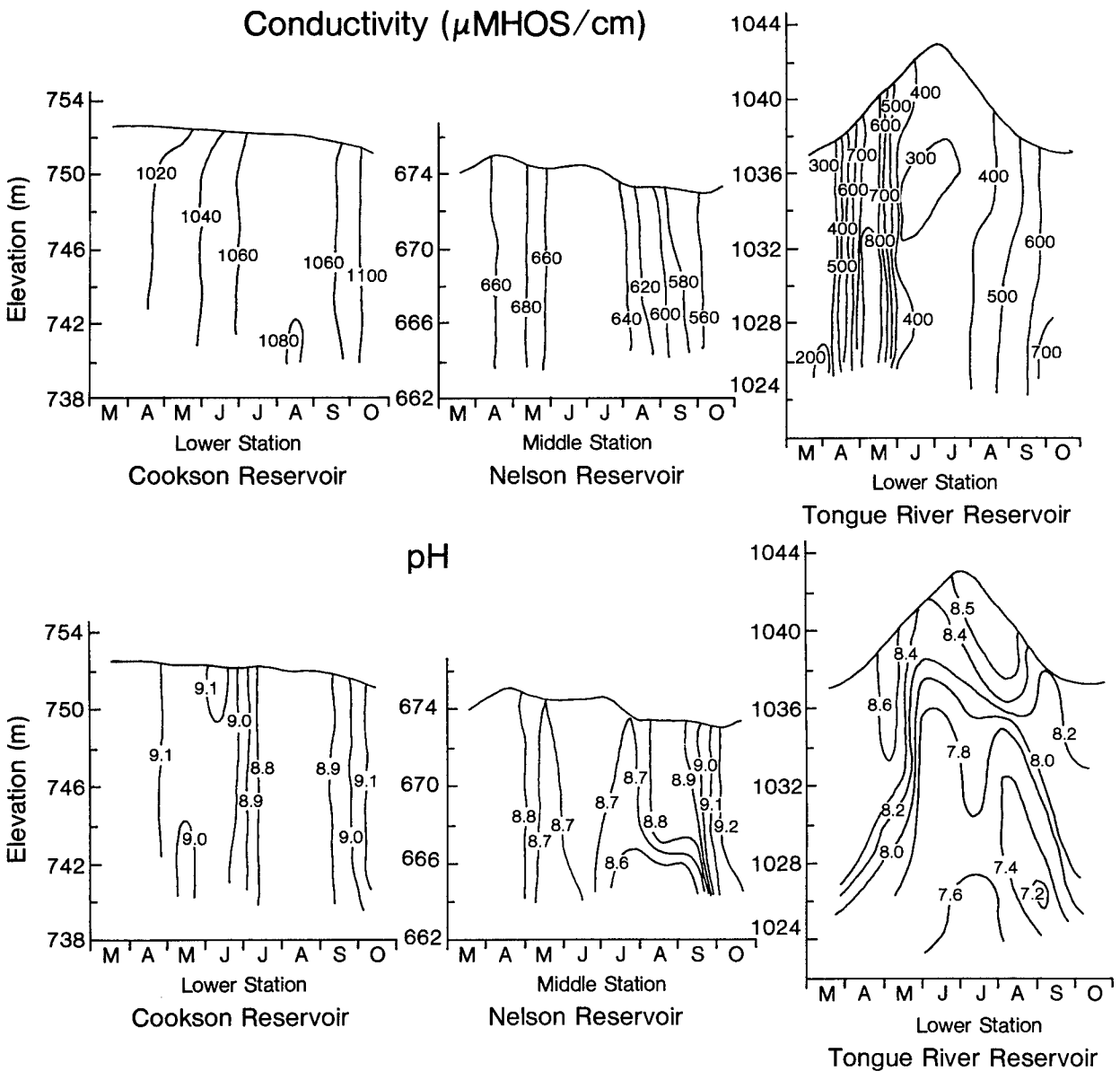

Fig. 35. Seasonal specific conductance (*upper*) and pH (*lower*) isopleths for the deepest station at Nelson, Cookson, and Tongue River reservoirs.

tosynthesis when carbon dioxide was being taken up. Carbon dioxide released by decompositional processes resulted in lower pH at the bottom in Tongue River Reservoir.

Mercury cycling is strongly influenced by pH. In acid freshwater lakes with about equal mercury inputs, low pH values correlated with higher methylmercury concentrations in fishes. Jernelöv and Åsell (1975) stated that higher methylmercury production occurs at lower pH because acid conditions result in the conversion of dimethylmercury to monomethylmercury. Additionally, more mercury binds to particulates upon acidification (Schindler et al. 1980), thereby preventing loss of mercury to the atmosphere. The rate of uptake of methylmercury by fish has a biphasic response to changing pH (deFreitas et al. 1977). At pH 5.5, methylmercury uptake was less than half that at pH 8.5, but was greater at pH 6.5 than at pH 7.5. Uptake was more rapid in hard water then in soft water.

Conditions favoring methylation of mercury and

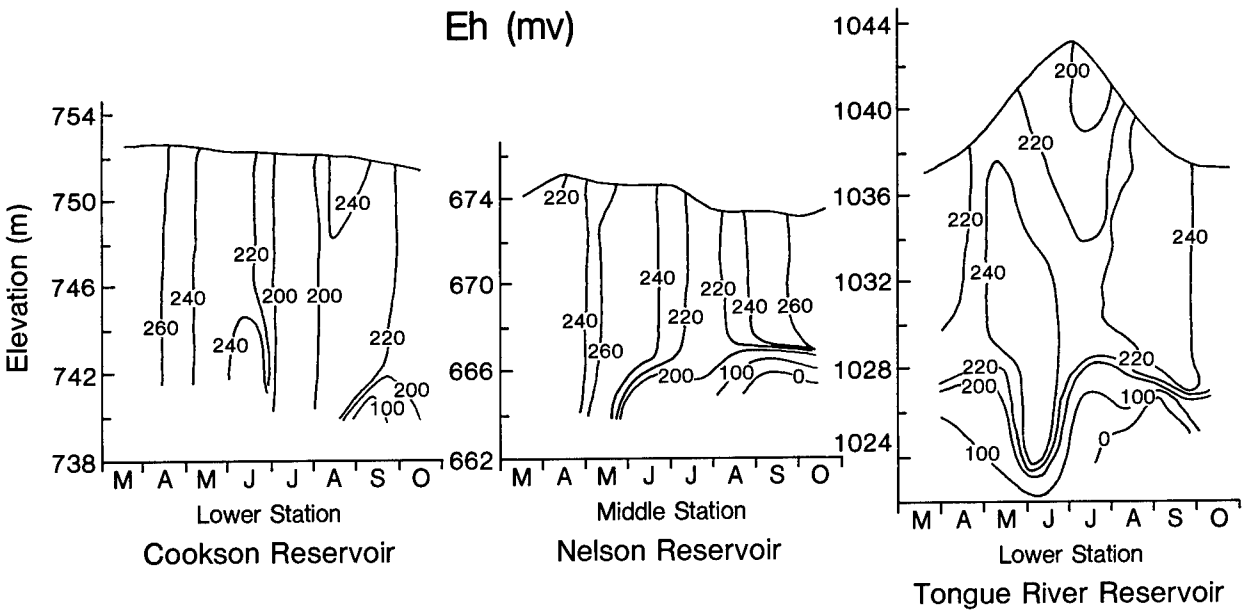

Fig. 36. Seasonal oxidative-reductive potential (E_h) isopleths for the deepest station at Nelson, Cookson, and Tongue River reservoirs.

methylmercury uptake by fish do not necessarily coincide. In the water column of our reservoirs, more dimethylmercury than monomethylmercury would tend to be formed because of the high pH (Bisogni and Lawrence 1975). Conversely, fish uptake of monomethylmercury would tend to increase with high pH and water hardness. Based on this line of logic, we would expect fishes in Cookson Reservoir to have the highest uptake rate, and those in Tongue River Reservoir to have the lowest; indeed this is what we observed.

Bacterial activity obviously increases as bacterial density increases; hence, bacterial density may be correlated with mercury methylation (Bisogni and Lawrence 1975). High bacterial densities observed in the water column of all three reservoirs during spring are due to an influx of nutrients from spring turnover and runoff. The slightly higher density of bacteria in Tongue River Reservoir (Table 25) appears favorable for methylation. However, mercury concentrations were lowest in fish from Tongue River Reservoir. Possibly the higher turnover rate in Tongue River Reservoir results in methylmercury being flushed downstream. The fourfold greater bacteria concentrations in the inflow to Cookson Reservoir than in the outflow may indicate high methylation rates and a high concentration of methylmercury in the stream entering the reservoir. Higher mercury concentrations in fish of Tongue River Reservoir during a flood year indicated that a significant portion of the methylmercury in reservoirs may originate upstream.

Conclusions

Total mercury concentrations in surficial sediments from the 10 reservoirs studied were uniformly low. Total mercury in sediments was not correlated with concentrations of mercury in fish; instead, mercury uptake by fish was correlated with variables that either facilitate mercury uptake or promote methylation.

Mercury and selenium in sediments from Missouri River Basin reservoirs were positively correlated with depth, presumably because deeper waters contain finer, more highly organic sediments.

Sediments from downstream Lake Francis Case and Lewis and Clark Lake contained extremely high concentrations of selenium, which appeared to originate in the White River drainage.

Fish from all of the reservoirs accumulated mercury logarithmically relative to fish length because growth in length slowed with age while mercury uptake rate remained constant.

Mercury uptake by walleyes from the reservoirs was sequentially related to position of the reservoir in the watershed (fish from upstream reservoirs accumulated mercury the fastest) and turbidity was strongly correlated with mercury uptake rates. Upstream reservoirs had less controlled inflows and more severe flood events that increased turbidity and scoured mercury and nutrients from terrestrial and riverine sediments. The general effect was to provide substrates and nutrients for bacterial growth in the presence of mercury, presumably stimulating methylation and also increasing the influx of methylmercury of terrestrial origin.

Conductivity, total dissolved solids, nonfilterable solids, and pH were all positively correlated with rates of mercury uptake by fish. The lower proportion of mercury present as monomethylmercury at higher pH may be offset by a higher rate of mercury uptake by fish.

Mercury content was considerably lower in fish from reservoir tailwaters than in reservoir fish of the same species and size. This is evidence that the reservoirs promoted mercury uptake by fish, either by increasing the amount of methylmercury available to fish or by providing conditions that stimulated methylmercury uptake.

In Tongue River Reservoir, mercury concentrations in northern pike were significantly higher in a year following a severe flood than in earlier or later years. Flooding was an important factor in mobilization and bioavailability of mercury.

Mercury concentrations were significantly higher in walleyes from Cookson Reservoir, the only new reservoir in our study area, than in walleyes from any of the other reservoirs. The inundation of terrestrial soils that occurs when a new reservoir is filled seems to create conditions that promote mobilization and subsequent bioavailability of methylmercury.

Most of the mercury transport into Tongue River Reservoir was by river water (93%); point sources from mining accounted for about 1% of this mercury, and the Sheridan, Wyoming, sewage treatment plant contributed 9%. The rest of the river input was probably a result of natural weathering. For sources of mercury other than surface runoff, we estimated that groundwater contributed 0.02%, precipitation 4.5% and dry deposition (including that from mining) 0.1%. Mining was not indicated as a significant point source of mercury to Tongue River Reservoir.

During 1980, more mercury left Tongue River Reservoir than entered it; however, estimates for other years indicated that inflow and outflow volumes of mercury varied considerably from year to year. Although we made calculations for only 3 years, it appeared that the amount of mercury entering the reservoir increased during years of flooding.

In 1980, Tongue River Reservoir at full pool contained 1.47×10^3 g of mercury—only about one-third of the quantity that entered and left the reservoir duing the year. These data show that the reservoir was a highly dynamic system with respect to mercury, and that yearly changes in mercury inputs substantially change the exposure of organisms to mercury.

Walleyes in Tongue River Reservoir, like those in most waters, were predominantly piscivorous, feeding principally on young-of-the-year white crappies. Invertebrates (principally chironomids) were eaten by young walleyes in spring.

The average and maximum length of fish eaten increased with walleye size; however, the minimum size of fish eaten changed little because young-of-the-year fish were eaten whenever they were available. Zooplankton and aquatic insects were prominent in the diets of white crappies from Tongue River Reservoir in April and June, but fish, especially young-of-the-year crappies, were prevalent in white crappie diets from August through October; crappies longer than 270 mm fed mainly on fish throughout the year.

White crappie diets varied diurnally; invertebrates were eaten primarily during daylight, and fish consumption increased at night. Daily feeding of white crappies in Tongue River Reservoir generally peaked at dawn, midday, and shortly after dark. Midday peaks were associated with consumption of invertebrates, whereas dawn and evening peaks corresponded with fish consumption.

Both walleyes and white crappies in Tongue River Reservoir fed opportunistically on white crappies, the most abundant forage fish present; both species appeared to select young-of-the-year fish when they were available. Annual food-consumption rates were estimated at 1.5–2.2% body weight per day for walleyes and 1.1–3.5% of body weight per day for white crappies.

Concentrations of mercury (μg Hg/g) averaged 0.08 (range, 0.02–0.40) in forage organisms; calculated average concentrations of methylmercury (μg MeHg/g) in fish diets averaged 0.05 for walleyes and 0.04 for white crappies.

Mercury concentrations (μg Hg/g) ranged from 0.02 to 1.22 in walleyes and from 0.02 to 0.53 in white crappies from Tongue River Reservoir; U.S. Food and Drug Administration consumption guidelines of 1.0 μ/g Hg/g were exceeded in only 2 of the 163 walleyes and in none of 248 crappies tested. Mercury concentrations in both walleyes and white crappies increased with increasing fish length and were higher in walleyes than in crappies of the same estimated age. This difference appeared to stem from differences in the amount of methylmercury ingested. As judged by our data, it seems highly unlikely that anyone would catch and eat enough walleyes or crappies to endanger their health.

The percent of accumulated methylmercury derived from food was estimated to be 41–62 for walleyes and 51–73 for white crappies; however, the error associated with these estimates is potentially large. Nevertheless, under conditions that can reasonably be assumed to occur, food was shown to be a major source of accumulated methylmercury in Tongue River Reservoir fishes.

Mercury methylation rates are likely to be high in all three reservoirs—Tongue River, Cookson, and Nelson—because thermal stratification is weak and nutrient concentrations that stimulate bacterial activity are high. Moreover the higher hardness and pH in Cookson Reservoir probably stimulate methylmercury uptake by pelagic fishes. The lower percentage of clay in sediments of Cookson Reservoir, along with the wind-generated mixing that produces aerated bottom waters, probably provides conditions that favor net methylmercury flux into the water column.

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